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Review

Lithium Ion Battery Models and Parameter Identification Techniques

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Abstract: Nowadays, battery storage systems are very important in both stationary and mobile applications. In particular, lithium ion batteries are a good and promising solution because of their high power and energy densities. The modeling of these devices is very crucial to correctly predict their state of charge (SoC) and state of health (SoH). The literature shows that numerous battery models and parameters estimation techniques have been developed and proposed. Moreover, surveys on their electric, thermal, and aging modeling are also reported. This paper presents a more complete overview of the different proposed battery models and estimation techniques. In particular, a method for classifying the proposed models based on their approaches is proposed. For this classification, the models are divided in three categories: mathematical models, physical models, and circuit models.

Keywords: battery modeling; lithium ion battery; storage system; parameter estimation

1. Introduction

Today, electrochemical battery systems are very important storage devices. Among them, lithium ion batteries represent a more recent and very promising technology. These batteries are used in both stationary and mobile applications because of their high power density and high energy density. Fully electric and hybrid electric vehicles are among the most important reasons for developing battery models and estimation technique that can predict the electric, thermal, and aging behavior of these storage systems under different working conditions, including various currents and temperatures. In fact, correct estimations of the state of charge (SoC) and state of health (SoH) are vital for a good electric vehicle range prediction and lifetime prediction.

An enormous quantity of research can be found in the literature on the development of different models with different levels of accuracy and complexity. Moreover, depending on the phenomena being modeled and the approach used to build the model itself, the reader might become confused by the different battery models and categorizations. Several surveys on these battery storage systems have been conducted for the different modeled aspects such as electric [1–4], thermal [5,6], and aging [7] factors. Starting from these surveys and other more recent works, this paper presents a more complete overview of all the above mentioned aspects and parameter estimation techniques.

The paper is structured as follows: in Section 2, an overview of several electric, thermal, and aging battery models divided into those with mathematical, physical, and circuital approaches is given. In Section 3, an overview of the different parameter estimation techniques divided into online, offline, and analytical approaches is presented. Finally, in Section 4, some conclusions with two simple summarizing tables are derived.

2. Battery Models

The literature contains much research on the modeling of lithium ion batteries. These models can refer to a certain physical aspect such as electrical, thermal, or aging aspects, or to a mixture of these. The electric models aim to reproduce, as well as possible, the behavior of the electric quantities (voltage, current, SoC...) at the external terminals of the battery. In contrast, the thermal models aim to reproduce the behavior and distribution of the temperature in and on the battery cell, in a one-, two-, or three-dimensional way. Finally, the aging models aim to reproduce the degradation of the battery, which is reflected by a capacity fade and/or internal impedance rise. Usually, this degradation is due to two different phenomena: calendar aging and cycle aging. The former is related to the degradation of the battery storage under certain conditions such as the temperature and SoC; the latter is related to the degradation of the battery as a result of cycling with a certain current profile and under certain conditions such as the temperature and SoC. Of course, these models can be combined to obtain models that are more complex but, simultaneously, more accurate.

Moreover, for almost all these aspects, it is common to base their modeling on a mathematical, psychical, or circuital approach. In the authors' opinion, every approach is a mathematical one. In fact, the physical and circuital approaches are based on differential equations and/or mathematical functions. On the other hand, the circuit and mathematical models refer to physical aspects, although, in a simplified way. What changes is the method used to model the physical system and its complexity and accuracy. Nevertheless, in order to be consistent with the previous works, we use the term "mathematical approach" for models that are described by a few simple equations, analytical or stochastic functions that try to reproduce the external behavior of the system without exploiting the real physical principles underling the system itself. They represent a higher level of abstraction of the physical system. We use the term "physical approach" for models described by differential algebraic equations (DAEs) or partial differential equations (PDEs) related to the real physical phenomena occurring in the real system. They are models that can use lumped parameters or distributed parameters in a one-, two-, three-, or four-dimensional way and represent the most detailed, accurate but, at the same time, complex models. Finally, when the system is modeled under simplifying assumptions in a one-dimensional way, through lumped parameters and using circuital elements, in particular for the electrical and thermal aspects, it is possible to use what we call a "circuital approach." This kind of modeling represents a method that falls between the other two approaches.

Therefore, in the literature, different organizations can be retrieved. In this paper, seeking clarity, we first divide the lithium ion models based on the approach used to obtain the model itself. Then, we subdivide each of these approaches based on the modeled aspect, as previously mentioned.

2.1. Mathematical Models

As previously mentioned, mathematical models can be divided into two main subgroups: analytical and stochastic models. In particular, it is possible to collect some electric and aging models from the literature based on these approaches.

2.1.1. Electric Models

Peukert's law [8] is one of the simplest analytical functions used to predict the capacity of a battery:

$$C = I^b t \tag{1}$$

where C is the battery capacity in Ah, I is the discharge current in A, b is the Peukert constant, and t is the time in hours. For an ideal battery, the Peukert constant is equal to one. For real batteries, however, it is between 1.2 and 1.7. For a constant discharging current, the results obtained using this function are rather good. In contrast, when the discharging current is not constant, this model fails.

Rakhmatov et al. [9–11] proposed both an extension of the Peukert model that also works when the discharging currents are not constant and a novel analytical battery model based on the diffusion

process of the active material into the battery. Furthermore, this proposed model is suitable for optimization algorithms that aim to maximize the discharging process. A comparison between the extended Peukert model and the proposed model shows that the latter gives results that are more accurate in terms of both the maximum and average errors. This comparison was performed using the results obtained through the Dualfoil battery simulation tool as reference results.

Another famous analytical electric model is the kinetic battery model [12]. This model is based on the kinetics of the chemical processes. As shown in Figure 1, the battery is modeled as two tanks.

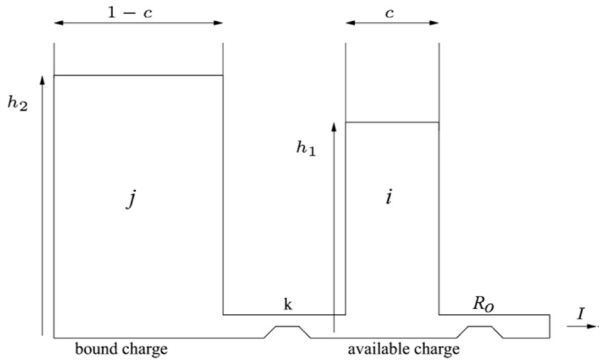

Figure 1. Kinetic battery model [12].

The total amount of liquid in these two tanks is related to the total SoC of the battery. The available tank contains the charge that is directly exchanged with the load. In contrast, the bound tank supplies the available tank through a valve with coefficient k. The speed at which the charge flows from the bound to the available tank depends on fraction c and coefficient k. The differential equations that describe the quantity of charge in the two tanks are as follows:

$$\begin{array}{l} \frac{di}{dt} = -I + k(h_2 - h_1), \quad i = h_1 c \\ \frac{dj}{dt} = -k(h_2 - h_1), \quad j = h_2(1-c) \end{array} \tag{2}$$

Chiasserini and Rao proposed [13–16] some of the first stochastic models. These models are based on the discrete-time Markov chain (Figure 2). Through the Markov process, it is possible to predict a future step of the system when only its present state is known. The history of the system is not needed. Thus, this kind of process is also called a memoryless process.

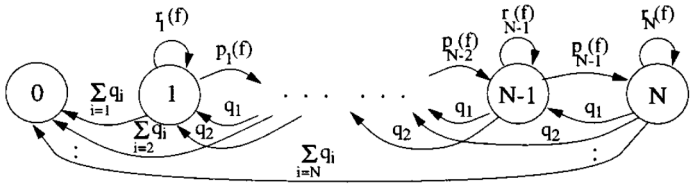

Figure 2. Markov chain of the stochastic process modeling the cell behavior [15].

In [13], two battery models for mobile communications are proposed. The first is simpler and is based on a discrete Markov chain with N + 1 states. Each of these states corresponds to the total charge available in the battery. In this model, for every time step, only one charge unit can be consumed with a certain probability q or, conversely, one charge unit can be recovered with the complementary probability $1 - q$. In addition, in the second model, more than one charge unit can be consumed at the same time step. Furthermore, it introduces a not nil probability of remaining in the same state.

In [14–16], the authors proposed other extensions of the previous stochastic battery models. In these cases, the charge recovery effect is also state dependent. In fact, if the total charge available

decreases, the probability of the recovery effect will be smaller. Moreover, these proposed models make it possible to develop different categories of protocols for energy efficient communications.

Rao et al. [17] repurposed the kinetic battery model in a stochastic way. In this case, a three-dimensional Markov process was used. Figure 3 shows the transition diagram of the proposed model, which has three state parameters. The parameters i and j are the same as those of Figure 1. They are related to the discrete quantities of the charge available in the two tanks, and the third parameters t is the time. Each transition between two different states is related to a certain occurrence probability. Furthermore, this stochastic model also considers the probability that the recovery effect cannot take place. Finally, simulation results proved that the model is fast with a good accuracy.

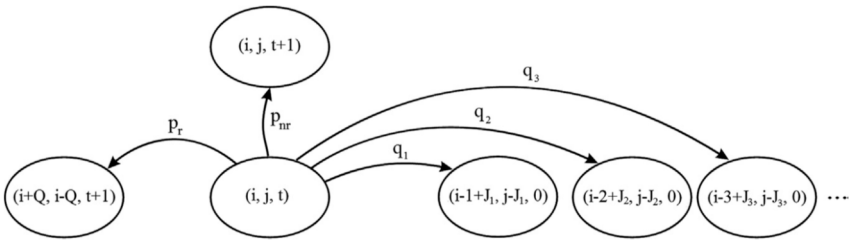

Figure 3. Transition diagram of the proposed model [17].

2.1.2. Thermal Models

One of the first mathematical thermal models able to predict the heat generation produced by a battery was proposed by Bernardi et al. [18]. The mathematical equation is as follows:

$$\frac{dq}{dt} = I(V - V_o - T\frac{\partial V}{\partial T}) \tag{3}$$

where I is the battery current, V is the internal battery voltage, V_o is the battery output voltage, T is the temperature, and q is the heat generated by the battery. Equation (3) is the most common expression because it is simple and, at the same time, takes into account the two important causes of heat generation: the joule effect, $I(V - V_o)$, and entropy change, $-T\frac{\partial V}{\partial T}$. On the other hand, it is not suitable for large batteries because it is based on the hypothesis that the current into the battery cell is uniform.

In [19], in order to extend the heat generation to larger batteries, an improved model was proposed that takes into account the current distribution into the battery itself. In fact, in the proposed model, the product between the specific area of the battery and the current density substitutes for the current term. The current density was calculated according to the polarization expression used by Tiedemann and Newman [20]. Finally, in [21], the heat generation due to the joule effect was expressed as a function of the electrical conductivity, and the entropy change was expressed as a function of the Gibbs energy.

2.1.3. Aging Models

These models can independently deal with the calendar aging, cycling aging, or both. In particular, the calendar aging seems to be well described by the Arrhenius equation, which considers the temperature as follows:

$$t = Ae^{-\frac{E}{RT}} \tag{4}$$

where t is the time-life, A is a constant, R is the gas constant, E is the activation energy, and T is the absolute temperature [22–25].

In [26], using the Taffel equation, which establishes a relation between the current flowing into the battery and its over-potential, the authors introduced the SoC in the calendar aging phenomena.

Many research papers agree on the fact that the calendar aging depends on the square root of the time as in [23], where an analytical approach was used and, as in [27], where, instead, a stochastic approach was used. Ecker et al. [28] developed a calendar aging model in which the behavior of both the capacity and the resistance of the battery are functions of the square root of the time. Simulations showed good agreement between the experimental and modeled results.

In [29], the authors proposed a calendar aging model based on the Eyring law instead of the Arrenius law. While the Arrenius law is an empirical expression, the Eyring law arises from the statistical thermodynamics and takes into account more factors than the Arrenius law. Indeed, the latter is a function only of the temperature, whereas the former, proposed in [29], also takes into account the SoC. The capacity fade of lithium ion batteries can be modeled using this law.

In [30], the authors performed several tests at different temperatures and SoC values, modeling the internal resistance increase. They derived a mathematical function according to which the internal resistance increases exponentially as a function of the temperature and SoC, and the time is elevated to 0.8.

Cycle aging models take into account the current in addition to the temperature and SoC, resulting in a much more complex calendar aging model. In this case, many works agree with the fact that, at least under certain temperature and SoC conditions, the cycle aging phenomena are related to the square root of the cycle number or, in other words, the charge exchanged with the battery [31–33]. In particular, in [34] the authors proved that, under fixed current and temperature conditions, the cycle aging does not depend on the duty cycle.

Other works, such as [35–38] estimate the aging of the battery by considering both the calendar and cycle phenomena together. Other mathematical models can be based on empirical data. In [39], the sum of the exponential functions and a regression analysis are used to model the aging of batteries. In [40,41], the authors proposed aging models based on the common method called "Coulomb counting." Fuzzy logic is used in [42–44] to model the aging with lower noise in the processed data. The electrochemical impedance spectroscopy (EIS) is another method used in [45] to find the amplitude and phase of the battery impedance as a function of the cycle aging.

2.2. Physical Models

Physical models are based on descriptions of the real physical phenomena occurring in the batteries. This typology is the most complex because it models, in an accurate and deep way, the electrochemical dynamics of the system using nonlinear equations.

2.2.1. Electric Models

One of the most important approaches for modeling the electrochemical processes of batteries is the lumped parameters approach based on a set of DAEs. This approach has been useful for modeling lead-acid and NiMH batteries. Conversely, it is not suitable to reproduce the more complex electrochemical behavior of lithium ion batteries.

A milestone for modeling lithium ion batteries was laid by Newman et al., who developed the porous electrode theory [46–49]. In fact, most of today's physical electric models are based on the principles of porous electrodes and a concentration solution. These models reproduce in detail the most important electrochemical processes such as the diffusion, mass transportation, and ion distribution using the PDEs. These latter are based on the diffusion behavior of the active material concentration (Fick's law [50]), the electrical potential distributions (Ohm's law), and the Nernst and Butler-Volmer equations [51]. Furthermore, many of these models describe the mass transportation and diffusion process using a one-dimensional spatial distribution [52–55].

Because these models are very complicated to simulate in real time, other more recent works have tried to reduce the order of these models using well-known model reduction methods, in order to obtain models that are less computationally expensive with or without a loss of information. For instance, Subramanian et al. [56–59] proposed several model reduction methods using perturbation techniques

and heuristic simplifications. The main drawback of these methods is the fact that it is necessary to preprocess the system in order to obtain a knowledge of its behavior under certain conditions.

In [60], the authors used the Chebyshev polynomial method, where the behavior of the state variables was reproduced in an approximate way through a linear combination of several Chebyshev polynomials.

Smith et al. [61,62] proposed a one-dimensional battery model based on another model reduction method called the residue-grouping method.

In [63,64], the authors proposed an extension of the model proposed by Smith et al., using analytical transfer functions to describe the concentration of the electrolyte and, later, developed a discrete time model based on transcendent transfer functions converted into a state-space model. These models have an important drawback because they work only for a fixed point of the SoC.

Conversely, Lee et al. [65] developed another model based on the blending approach that is able to work under a wide range of temperature and SoC conditions. Other reduction methods found in the literature include the proper orthogonal decomposition method [66], Padè approximation method [67], and Galerkin method [68].

Another more recent technique used to reduce the order of the models was based on the reformulation of the lithium ion battery equations as a state-space model [69]. All of the above reduction methods lead to battery models that are rather fast and, at the same time, much accurate practically without a loss of information. For applications in which less accuracy over a small working range is required, other faster electric physical models with a loss of information have been proposed [70,71].

2.2.2. Thermal Models

The most important physical thermal models use the distributed parameters in a one-, two-, or three-dimensional way. In any case, it is possible to derive a general expression that combines the heat generation with the temperature spatial distribution:

$$\rho\, c \frac{\partial T}{\partial t} - \frac{dq_s}{dt} = \lambda_x \frac{\partial^2 T}{\partial x^2} + \lambda_y \frac{\partial^2 T}{\partial y^2} + \lambda_z \frac{\partial^2 T}{\partial z^2} \tag{5}$$

where ρ is the density of the battery, c is the specific heat capacity of the battery, the $\lambda_{x,y,z}$ terms are the thermal conductivity in the x, y, and z directions, and q_s is the heat generated per volume unit.

In [72], based on Equation (5), a one-dimensional model was proposed and tested under different working conditions. In contrast, in [73], the authors proposed both a two-dimensional thermal model for small cylindrical batteries and a three-dimensional thermal model for larger prismatic batteries.

In [74], the authors developed a thermal model that can work under different charge currents, particularly for quick charging EVs. This model takes into account the current distribution into the battery cell.

In [75], another thermal model was proposed. In this case, in addition to the current distribution, other factors were taken into account, such as the local concentration of the lithium.

One of the most recent works was reported in [76]. In this work, the authors proposed a thermal model that used the so-called isometric feature mapping (ISOMAP) method, which is a nonlinear reduction method. In other works, different distributed thermal models have been coupled with an electric-chemical one yielding electro-thermal models [77–79].

2.2.3. Aging Models

As for the physical electric models, the first kind of aging model was born from the work of Newman et al., the first attempt was done in [80], in which a solvent oxidation reaction was implemented. The choice to use the solvent model was very promising for the next works. In fact, in [81,82], Christensen et al., using the solvent model, proposed an aging model explaining both the capacity fade and the resistance increase of batteries due to the formation of the solid-electrolyte

interphase (SEI). Another study on the SEI formation was made in [82]. Other important research works, as in [83], developed models able to simulate the composition of the electrolyte and the evolution of the battery performances as a function of the cycle number. Ramadass et al. [84] developed a model that also takes into account the side reactions on the negative electrode of a lithium ion battery.

According to the kind of electrodes used in the lithium ion battery, different chemical phenomena can take place. Based on the particular kind of phenomenon to be modeled, different investigation methods have been developed. For instance, in [85], the authors modeled the aging behavior of a LiCo02 battery by performing a so-called ab initio calculation, while, in [86], a study was conducted to understand some processes of LFP batteries. Afterward, Tasaki et al. [87,88] studied the lithium salt dissolution near the SEI of the negative electrode, obtaining a link with the capacity fade due to the calendar aging. Regarding graphite electrodes, in [89], the authors analyzed the decomposition of the solvent related to the SEI formation. In [90], the SEI evolution was studied and related to the decrease in the electrode active material due to cycling.

Another category of simplified physical aging models is called the single particle model. These are very accurate only for low discharge currents. In [91], the current distribution was assumed to be constant, although the electrodes and dynamics of the electrolyte were ignored. In order to overcome the problem of low accuracy at high currents, different methods have been proposed. In [92], it was demonstrated that the single particle model is not accurate for high currents as a result of ignoring the dynamics of the electrolyte. Therefore, in [93], the Laplace transform method was proposed to describe the dynamics of the electrolyte using analytical transfer functions. In contrast, in [94], an integral method was proposed. Other more recent methods to model the electrolyte concentration were reported in [95,96].

2.3. Circuit Models

2.3.1. Electric Models

Electric circuit models are a good solution in terms of the tradeoff between the mathematical models and the physical ones. In fact, they are less complex than the physical models and, at the same time, they present a medium accuracy. Moreover, they are simple to implement for real time applications.

The simplest electric model is composed of an ideal voltage source without representing the battery's internal characteristics, as reported in Figure 4a. A next step is to add a resistor in series with the voltage generator to represent the internal voltage drop, as shown in Figure 4b. These two parameters are constant and do not depend on the SoC [97]. Of course, these models are not good at describing the charge/discharge voltage profiles. At the same time, these models can only be used in applications where the energy of the battery can be supposed to be infinite.

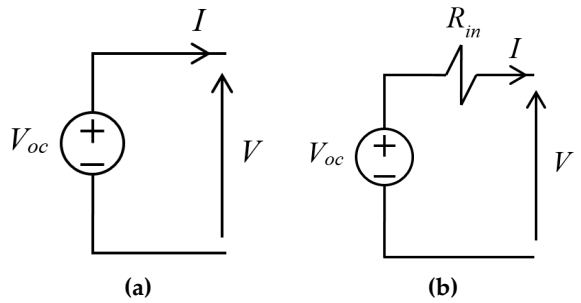

Figure 4. Simplified electric models: (**a**) ideal voltage source; (**b**) real voltage source.

In [97–99], both the internal resistance and voltage source are functions of the SoC. These models start to be more accurate at representing the charge/discharge voltage profiles. Different characteristics

can be derived according to the functions used to represent the voltage source and internal resistance. These models are called simplified models.

So far, these electric models have been static models because the dynamic behavior of the battery has not been considered. On the other hand, electric transients cannot be modeled. Another important category of electric model, which also considers the dynamic behavior, is the so-called Thevenin model. These models are composed, as the previous ones, on a voltage source in series with the internal resistance, along with different RC parallel branches. These latter are used to reproduce the dynamic behavior of the battery, as reported in Figure 5. A larger number of RC branches results in a better accuracy for the reproduced dynamic behavior.

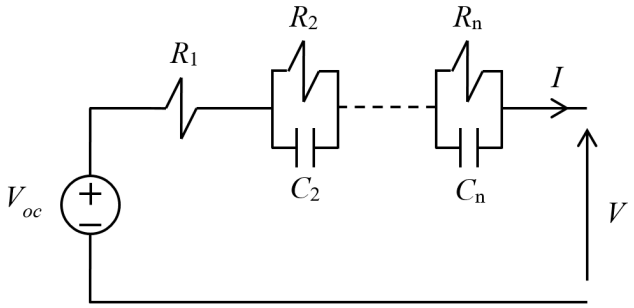

Figure 5. Thevenin electric model.

In [100], the simplest Thevenin model, in which only one RC branch is used and all the parameters are constant, is reported. The latter point is the major drawback of this model. To overcome this issue, the same model can be improved by considering that all the parameters, or only a subset, are dependent on different factors. For instance, in [101–103], the SoC dependency is taken into account. In other works, such as in [104–107], the temperature and/or capacity fade are also considered.

The simplified models represent a subset of the Thevenin models. For this reason, it is possible to categorize all these models as a unique group, where Figure 5 represents the most general case. According to the accuracy and based on the aim of the application, it is possible to choose a different number of RC branches. When the application needs to represent only the steady state behavior, the model becomes one of the two reported in Figure 4. In any case, the number of RC branches needs to represent the transient behavior as well as possible. The accuracy of the behavior of the output battery voltage depends on which parameters are dependent. In other words, it is dependent on the model variables. In this way, the same model can be more or less complex according to the parameter dependency. In fact, the same model can have parameters that depend on one or more of the following factors: the SoC, current, temperature, SoH, and so on, as follows:

$$p_i = f_i(\text{SoC}, \text{SoH}, T, I) \tag{6}$$

where p_i is the i-th parameter of the model (V_{oc}, R_1, C_1,...).

In the literature, it is possible to find another category of electric circuit models called impedance models. These models are very similar to the Thevenin models. The difference is the technique used to estimate the parameters. Nevertheless, one of the most important so-called impedance models is based on the well-known Randless equivalent circuit model [108]. It is used to describe a half-cell of a battery.

The model proposed in [109] based on the Randless equivalent circuit model is shown in Figure 6. This model is composed of resistor R_{in}, which is related to the internal resistance of the electrodes, electrolyte, and separator; capacitor C_{d}, which takes into account the double layer capacitance effect; resistor R_{SEI} and capacitor C_{SEI}, which are the resistance and capacitance of the solid state interface layer; resistor R_{ct}, which is related to the charge transfer effect; and, finally, the Warburg impedance

Z_W, which considers the diffusion process. This electric circuit is completed by adding, as done for the previous models, the internal voltage source V_{oc}.

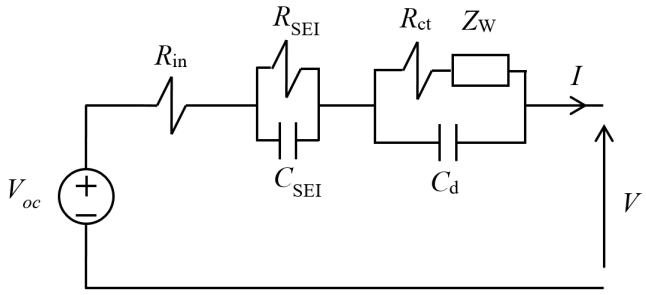

Figure 6. Impedance electric model based on Randless circuit.

The Warburg impedance is defined as follows:

$$Z_W = R\frac{\tanh(\sqrt{j\omega C})}{\sqrt{j\omega C}} \tag{7}$$

where R and C are the two parameters characterizing the Warburg impedance itself.

In [110], the Warburg impedance is moved into series with the other parallel branches, as reported in Figure 7, and the R_{SEI} C_{SEI} parallel branch is also replaced by the so-called ZARC element. The latter is composed of a resistor in parallel with a constant phase element (CPE) [111]. The CPE is a generalized capacity defined as follows:

$$\text{CPE} = \frac{1}{(j\omega)^{\phi}C} \tag{8}$$

where C is the capacity, and φ is the depression factor. Therefore, the ZARC element based on the depression factor is a generalized RC parallel branch. If this factor is equal to one, the CPE becomes a classic capacitance, and the ZARC element becomes a classical RC branch.

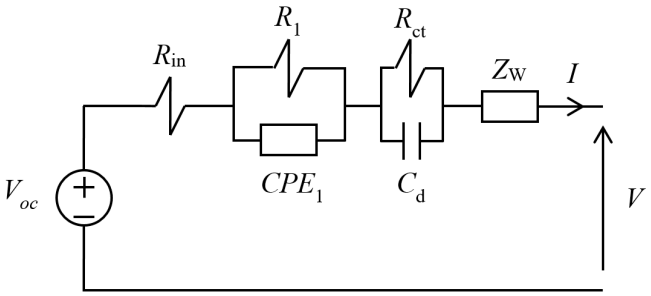

Figure 7. Generalized impedance electric model.

In general, it is possible to build an electric circuit model with a different number of the above mentioned elements (ZARC and Warburg elements) according to the chemical phenomena that occur in the battery [112]. In any case, all the parameters can be functions of different factors, as previously discussed for the Thevenin models.

Furthermore, both the ZARC elements [113] and Warburg impedances [114] can be approximated by the sum of a variable number of RC parallel branches. In practice, it is sufficient to use a few of these RC branches to have a good approximation. In this way, the so-called impedance electric models become Thevenin models with a large number of RC branches. Therefore, it is possible to consider the model of Figure 5 as the most general one.

2.3.2. Thermal Models

If the temperature is assumed to be uniform or piecewise uniform in the battery, it is possible to use lumped elements to describe the thermal behavior of the battery itself. As done for the electric circuit, in this case, the same elements can be used to represent thermal resistances (the inverse of the thermal conductivities), thermal capacitances, and heat sources. Therefore, it is possible to solve the thermal circuit, finding the temperatures inside and on the cells, by applying the same rules of the electric circuit theory (Figure 8).

The authors in [115], using the energy balance given by Bernardi et al. [18], developed a one-dimensional model that could predict the temperature of a lithium ion cell. Moreover, the proposed model could be used to simulate many kinds of separator materials. Simulation results at different temperatures and discharge currents are presented.

In [116], the authors proposed a one-dimensional model in which the decomposition of the anode is taken into account. In this way, it is possible to reproduce the temperature behavior of the cell as a function of the discharge conditions. The results showed that the size of the particles inside the anode is important for the temperature behavior.

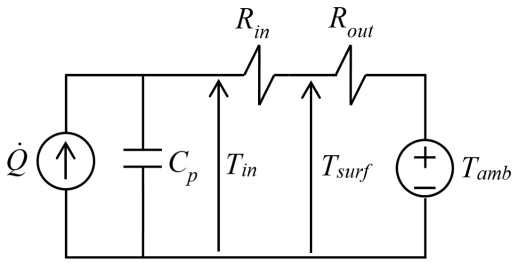

Figure 8. Lumped thermal model [117].

Forgez et al., in [117] developed a simple thermal model for a cylindrical lithium ion battery. In order to determine the parameters of the model, a thermocouple was inserted into the cell to measure the internal temperature. Then, with another thermocouple used to measure the temperature on the battery surface, they used the proposed model to estimate the internal temperature with an error of 1.5 °C. In [118], the model proposed by Forgez et al., was used and integrated with an electric model.

3. Parameter and State Identification Procedures

The models proposed in the technical literature differ not only in their chosen topology and type, but also in the techniques adopted to estimate both the parameters and states of the battery model. The term parameters refers to the characteristic quantities of the system, including chemical (solid phase conductivity, diffusion coefficients...) and electric quantities (internal resistance, capacitance...), while the states are the variables related to the system evolution, i.e., they have a knowledge of the history of the system, such as the SoC and SoH.

Some models are more accurate than others in predicting the battery behavior, but the procedures necessary to estimate both the parameters and states can be very difficult, limiting the usability of the model itself. For this reason, easier models are widely used because complex models require too much effort to be adopted. The necessary effort includes both human effort to set up and perform the necessary tests and consequent calculations, and hardware effort to implement the calculations. Therefore, different methods for optimizing different aspects (i.e., the simplicity, speed, accuracy, robustness, and reliability) have been proposed. In the rest of this section, an overview of the methods proposed to estimate the parameters and states of different models is reported.

In this overview, the classifications proposed for these models are not used. This is why the specific features of the parameter and/or state estimation methods are quite different from those characterizing the models. It is worth noting that, while many techniques have been proposed to

identify the necessary parameters for electrical models, a lower number of identification techniques is available in the literature for thermal and aging models.

The identification methods can be divided in three main categories:

- online identification methods;
- offline identification methods;
- analytical or numerical calculation methods.

While the first two are based on the analysis of the results of experimental tests, the last is obtained by means of analytical calculations directly derived from the physical principle at the basis of the battery. The first two are more diffused because they can also be implemented without a deep knowledge of the device and are not affected by the errors connected with the manufacturing process. The online identification methods are designed to allow parameter/state estimation during the normal operation of the battery, while the offline methods are developed by testing the batteries with ad-hoc tests realized when the battery is not used for its application. The main important characteristics that online methods have to guarantee are as follows:

(1) computational simplicity allowing real-time execution (possibly also as a background process);
(2) the capability of estimating all the parameters/states using only measurements obtainable by the hardware normally connected to the battery;
(3) the capability of estimating all the parameters/states using only the normal operating conditions of the battery itself.

In practice, many methods are mixed methods because they usually estimate states online and parameters offline by means of dedicated tests performed only before the installation in specific situations. This is frequently the case for the open circuit voltage (OCV), which is estimated when the battery is not operating over a long period (usually at least 30 min).

As previously mentioned, the third family of identification methods is based on analytical modeling or finite element calculations. These methods should be offline. However, because they are not based on the results of experimental tests, it is preferable to classify them separately. Indeed, the calculations could be repeated as functions of some measurement (i.e., current, voltage, or temperature) and could be used to tune online the parameters values. In this case, they could be used online.

In the following, online and offline methods will be analyzed separately, even if some of them are mixed methods. In the overview of the technical literature, all of the model types described in the previous section will be taken into account (i.e., electric, thermal, and aging).

3.1. Online Identification Methods

The majority of online identification methods are applied to equivalent circuits representing the electric behavior of the battery. In particular, the online methods usually focus on SoC estimation during the normal operation of the battery. Indeed, because the SoC varies continuously during battery operation, only an online estimation of the SoC is sound. Some or all of the other parameters depend on the SoC, and their estimation can be online or offline (as in mixed methods).

In recent years, several techniques have been proposed for SoC estimation. The basic method is Coulomb counting, which consists in the integration of the current. This method suffers from all the problems connected with the drift of the integral, in addition to the difficulty of estimating the initial SoC. In order to solve this problem, some studies have proposed the addition of a term obtained by PI regulators to the Coulomb counting [119,120]. Because the SoC is somehow connected to the OCV, several methods have attempted to estimate the OCV and then estimate the SoC on the basis of previously performed offline experimental tests. The OCV is usually estimated on the basis of the selected equivalent electric circuit. In this case, errors in the circuit parameter estimation influence the accuracy of the OVC and consequently the SoC determination. For this reason, the use of

extended Kalman filters (EKFs) to simultaneously estimate all the parameters together with the OCV is proposed in some papers [121,122]. In order to reduce the computational burden connected with the implementation of EKFs, in [123], an adaptive EKF was presented.

The online estimation of the OCV, using offline techniques for all the other parameters, was also considered in [124], which applied the Thevenin model (see Figure 5) with only one RC branch. For the parameter estimation, a set of pulsed charge/discharge tests were used, and a prediction error minimization algorithm was applied to the test results. Polynomial approximation was used to interpolate all the results with the 8th degree for the relationship between the OCV and SoC and 5th order for all the other parameters. The SoC was estimated online using an adaptive neuro-fuzzy inference method. This approach showed good accuracy, but the use of high order polynomials made the results strongly dependent on the experimental data and unstable with parameter variation. For this reason, the results are not portable to other cells, and all the batteries have to be identified to maintain good accuracy. On the same 1-RC branch model, in [125], an approach based on least square minimization (LSM) was used in the Laplace domain to identify all the parameters that were functions only of the SoC. This was estimated using a single OCV-SoC curve obtained by means of an offline test. The OCV-SoC curve did not take into account the temperature and aging effects, which limited the usability of this model. This problem was partially solved in [126], where an adaptive algorithm for parameter estimation on a model with two RC branches was proposed. In this paper, the parameters were obtained by means of offline discharge tests performed at different currents and temperatures. After estimating the SoC, the parameters were adjusted online. Moreover, a method to identify the SoH of the battery was proposed, but its value was not used to modify the parameters. Thus, the identification procedure took into account the temperature effect but not the battery aging. In addition, in [127], a circuit with two RC branches was taken into consideration. In this paper, an innovative definition of the SoC was given on the basis of a mechanical analogy. Parameter estimation was performed offline on the basis of a step changing current that was both positive and negative. In contrast, the new SoC was estimated online by means of an EKF. In addition, in this case, the temperature and aging were not taken into account, but the usability of the model was proven on more cells connected in series, and it showed a good stability in the portability of the parameters. In [128], only the OCV versus SoC relationship was taken into account. This relation was described by means of a sigmoid function written as the sum of a constant and four exponential terms. The online estimation of the SoC from the OCV was obtained by applying an EKF. The problem of estimating the OCV during normal operation was not taken into account in the paper.

Another set of parameter and state estimation methods is based on physical electrochemical models. Such methods are more accurate than the previously mentioned ones, yet are more complex. In particular, as discussed in the previous section, most of the physical electric models are based on the principles of porous electrodes and the concentration solution, and they have very complex mathematical structures. For this reason, other more recent works have proposed different reduction methods to make those models less complex and more suitable for developing parameter/state estimators. In [129], a linear Kalman filter was implemented to reduce the model proposed by Doyle et al. [47,52], in order to estimate both the current and voltage limits for a lithium ion cell under pulse operations. Starting from the measurements of the current and output voltage of the cell, the concentration gradient, battery potential, and SoC were estimated, through which the operation limits of the cell were also estimated. In [130], the authors proposed a state estimation procedure for a 1D reduced electrochemical model. In this case, the model used in the estimating procedure was more detailed than the previous one, and it was also possible to predict the internal temperature of the cell in addition to the concentration gradient, battery potential, and current density on the electrodes. This could be very useful if integrated into a battery management system for safety reasons and optimal battery utilization. In particular, this model made it possible to obtain a mathematical expression of the capacitance as a function of the voltage relaxation effect. In [131–136], other SoC estimation procedures were developed based on single particle models. In particular, in [131], the authors used

an EKF to estimate the SoC by considering the negative electrode of the lithium ion cell as the limiting electrode. Moreover, in [132], the authors implemented an EKF where the electrochemical model was reformulated as a linearly spatially interconnected system. In [133], the estimation of the SoC was carried out using the back-stepping observer for PDEs, and analyzing the observability proprieties of single particle models. In this way, the mathematical structure of the single particle model was greatly reduced, and as a result, a nonlinear, simple, and smart PDE observer was obtained. In [134], the same authors proposed the estimation of the SoH based on the swapping identification method for the parameters in the PDE of a single particle model. In [134], the authors developed two different procedures for SoC estimation, both based on the Luenberger observer. The first used the Luenberger observer with a constant gain, whereas the other used the output Jacobian as the weight of the Luenberger gain. In any case, the problem was turned into a linear matrix inequality to be solved through the Lyapunov method. An extended version of this work was reported in [136].

Unlike the above estimation methods, which were finalized to identify the SoC, SoH, and other state variables such as the internal temperature and battery potential, the so-called adaptive or state-parameter estimation methods represent another type of identification procedure for simultaneously estimating both the parameters and states. Important works can be retrieved in [137–143]. In these procedures, it was difficult to verify the convergence of the estimator. In particular, in [137], a multi-particle filter was applied to a full electrochemical model without any simplification of the mathematical equations, and both the parameters and states of the model were estimated using a Bayesian framework. In [138], the authors proposed a method to estimate both the residual power and capacity of a lithium ion battery using a lumped parameter model with an unscented Kalman filter state predictor. Two parameters are considered to be more sensitive to the aging phenomena and are estimated through the LSM approach. In [139,140], the authors used a single particle model to develop an adaptive SoC estimator, where the observability of the SoC was connected to the model parameters. Through the use of an iterated EKF, both the SoC and parameters were simultaneously estimated. In [141,142], the authors developed SoC estimators without using any linearization of the nonlinear model. In fact, in these works, nonlinear geometric observers were used to estimate both the parameters and SoC. Moreover, it was possible to verify the convergence of the estimator. In [143], the authors presented an estimation procedure based on three sub-observers, where two of them were adaptive sliding model observers, to identify the SoC and SoH in a combined way. In addition, in this case, the convergence verification of the estimators was possible using Lyapunov's stability theory.

None of the above reported electrochemical-based estimation methods considered the thermal effects. In this regard, the authors of [144] used a Luenberger observer in a single particle model to estimate the SoC by also taking into account the temperature and electrolyte effects. Conversely, the model parameters were not estimated. To fill this gap, in [145], the authors developed an adaptive observer procedure, applied to a coupled electrochemical-thermal model, through which it was possible to simultaneously estimate both the SoC and SoH, along with some model parameters. Lyapunov's stability theory was also used in this case to assure the convergence of the estimation procedure.

3.2. Offline Identification Methods

As previously discussed, several papers have proposed offline estimation methods for the parameters of an equivalent electric circuit and an online estimation method only for the SoC and SoH. The parameters of the equivalent electric circuit change according to the operating variables such as the actual SoC, SoH, current, voltage, and temperature (humidity is never considered because of the hermetic package of a commercial battery). Although the parameter characterization takes into account all of these factors, their estimation can obtain very high accuracy when done offline. Of course, the main advantage is the reduced complexity of the modeling activity and algorithm used for the online estimation of the SoC and, if necessary, the SoH. The main drawback is the difficulty of characterizing the parameter variation with all the factors, especially the SoH and temperature. The problem of defining the parameter variation with the SoH mainly depends on the duration and

difficulty of the tests to be performed. Indeed, during battery aging, the parameter variation, along with all the other factors, has to be evaluated until the end of life of the battery itself. Then, the test has to be repeated on a large number of cells to obtain reliable results. In contrast, the problem with characterizing the parameter variation with the temperature is the difficulty of controlling the temperature of the entire cell to a desired value. Indeed, the current flowing in the cell causes a non-uniform temperature distribution in the cell. Moreover, in many studies, the tests were performed in climate chamber, but this is not sufficient to fix the cell temperature, which changes according to the internal losses of the cell.

The majority of the papers propose offline procedures for parameter estimation based on different tests. Some papers propose complex procedures that achieve better results, while others prefer simple identification procedures and simple tests, even if the accuracy of the results is lower. In the following, starting from the simplest procedures, the identification techniques proposed in the literature will be quickly reported. In [146,147], with reference to an electrical equivalent model with only one RC branch (see Figure 5), the parameters were obtained using simple constant current discharge curves. Because the dependence of the model parameters on the current and temperature was considered, the tests were repeated at different temperatures and currents. The tests were performed in a climate chamber, but the temperature of the cell was not controlled. In [147], tests were also used to define an apparent state of discharge accounting only for the usable part of the residual energy inside the battery. In [124], for the same model, the resistive and capacitive parameters were estimated using pulsed discharge tests. For a model with two RC branches, in [148], different tests, at a constant current and temperature, were performed for the charge and discharge. This was done with the aim of identifying some hysteresis effects in the battery behavior. All of the tests were repeated at different temperatures to account for the parameter dependence on the temperature, while the effect of the current on the parameters was not considered. Again in this case, only the environmental temperature was controlled and not the battery temperature. Increasing the complexity, some studies [149–151] proposed the use of EIS tests to identify the parameters of 1-RC or 2-RC models by looking at the frequency response of the battery cell. The results were good, but the tests required dedicated hardware and the very slow response (time constants higher than hours) could not be taken into account. Thus, when increasing the time period of the sinusoidal current used for the EIS and working with very low currents, the moved charge became too high to consider a constant SoC. Moreover, the dependence of the parameters on the current could not be obtained by means of the EIS.

In [152], a more complex model, integrating the electrical and thermal behaviors, was considered. The electrical behavior was modeled with a 2-RC branch model, but the results were also repeated and compared with models using one or three RC branches. For the electrical parameter estimation, the pulse discharge was used at different temperatures. In this case, the temperature of the cell was not strictly controlled, but a flow chamber for cooling was used, and the temperature of the cell was monitored. The parameters of the thermal model were obtained by means of LSM, taking into account a cylindrical symmetry for cylindrical cells. The modeled variation of the resistances with the temperature was obtained by applying an Arrhenius model whose coefficients were identified using the tests results. For the two-dimensional thermal model presented in [153], a LSM method was used to estimate the parameters by considering the FEM results as tuning data. The procedure worked at a steady state. Therefore, a pulse current was simulated for a time long enough to have constant temperatures. In the estimation procedure, the losses were considered to be known, which implied a knowledge of electrical parameters independent of the temperature. Experimental validation was also proposed for this method. The problem of the dependence of the electrical parameters of the heating source was solved in [154], where a numerical method (similar to a LSM) was used to identify the parameters. In order to understand the heating source, a symmetrical (charge/discharge) pulse current test was arranged. In this way, the heating source could be evaluated by measuring the voltage and current without knowing the electrical parameters.

In the literature, some parameters are evaluated offline to identify the aging of a battery. In [128], the authors looked at the incremental capacity as an indicator of the aging. The incremental capacity was estimated using dedicated discharge curves at fixed currents and three different temperatures. The reduction of the incremental capacity was correlated with the battery SoH, while the effect of the temperature was only qualitatively reported. In [155], the incremental capacity was estimated for one temperature as an aging indicator. In addition, the impedance growth was also taken into account, but the effects were not quantified in parameters to identify the SoH variation. In [34], with reference to a simple model for which the available capacitance was only related to the moved charge, an experimental campaign was presented to estimate the aging coefficients. If the model was very simple, the necessary tests were very time consuming (months) and should be repeated for different cells/technologies.

3.3. Analytical Methods

As previously discussed, some models have been derived by studying the physical phenomena occurring in a battery. These models can also be used to calculate and predict the parameter values. The main advantage is the possibility of avoiding experimental tests to use the model. The main drawback is the uncertainty of the results connected with the impossibility of having a device behave exactly like the model. Indeed, real devices are different from their designs because of differences in the prime material and, mainly, as a result of causes connected with the manufacturing process. For particular applications, where all the geometrical and chemical parameters are well-known, analytical methods are used to obtain reference values for the proposed experimental procedures. This is the case in [156], where an offline procedure was proposed to estimate the thermal parameters of a battery model, and the results were compared with those obtained by means of calculations. These methods are more commonly used to design new cells than to characterize the behavior of existing devices. For this reason, they are more often developed for chemical models and can be found in specific electrochemical journals [157,158].

4. Conclusions

In recent years, batteries have become increasing widespread in many applications. Their power and energy range from a few watts and watt-hours to several megawatts and megawatt-hours. Among batteries, lithium-based batteries are the most promising technology because of their high power and energy densities. For this reason, in the last few decades, many models have been proposed to represent their behaviors.

This paper presented a broad overview of the methods proposed in the technical literature to model the behaviors of lithium-based batteries. In particular, the models were divided in three main categories according to their approach in modeling the battery: mathematical models, physical models, and equivalent circuits. For each category, papers on the electrical, thermal, and aging behaviors of the batteries were reviewed and quickly summarized. In the analysis of the proposed models, it was clear that many models are very similar, but they differ in the algorithms proposed to identify their parameters. For this reason, in the second part of the paper, the parameter identification procedures proposed in the literature were analyzed. They were divided into online, offline, and analytical procedures and a deep overview was presented. In order to make the proposed overview clearer and more useful to the readers, Tables 1 and 2 are presented in this section.

In Table 1, the lithium battery models available in the technical literature are summarized, highlighting their advantages and drawbacks and citing the papers where they were presented. In Table 2, the advantages and drawbacks of each family of methods for parameter estimation are reported with references to the papers using that method.

Table 1. Models.

Model	Advantages	Drawbacks	Papers
Mathematical	Very simple and low time consuming	Low accuracy	[8–17] [18,19,21] [22–45]
Physical	Very high accuracy	Very complex and time consuming	[46–49] [52–71] [72–79] [80–96]
Circuit	Simple and intuitive to be implemented	Medium accuracy	[97–110,112] [115–118]

Table 2. Parameter identification methods.

	Method	Advantages	Drawbacks	Papers
Online	Coulomb counting based	Simplicity	Drift of integral, tuning of regulators	[119,120]
	Observers	Very good accuracy, contemporary estimation of all the parameters	Complexity, time consuming	[121–123,127–145]
	LSM	Good accuracy	Medium complexity, dependency of the solution on the initial point	[125,126]
Offline	Constant current discharge	Standard instrumentation for testing	Impossibility of identifying eventual hysteresis phenomenon	[128,146–148]
	Pulsed charge/discharge	Analysis of charge and discharge operations	Medium complexity for implementation	[124,152–154]
	EIS	Information about frequency response, fast tests	Impossibility of identifying parameter dependency on current, necessity of dedicated hardware	[149–151,155]
Calculus	-	No need of tests and instrumentation	Low accuracy and impossibility of taking into account manufacturing effect	[156–158]

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Abbreviations

CPE	Constant Phase Element
DAE	Differential Algebraic Equations
EIS	Electrochemical Impedance Spectroscopy
EKF	Extended Kalman Filter
LFP	Lithium Iron Phosphate
LiCo02	Lithium Cobalt Oxide
LSM	Least Square Minimization
NiMH	Nickel Metal Hydride
OCV	Open Circuit Voltage
PDE	Partial Differential Equations
SEI	solid-electrolyte interphase
SoC	State of Charge
SoH	State of Health

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