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A Circular Economy of Electrochemical Energy Storage Systems: Critical Review of SOH/RUL Estimation Methods for Second-Life Batteries

Simon Montoya-Bedoya, Laura A. Sabogal-Moncada, Esteban Garcia-Tamayo and Hader V. Martínez-Tejada

Abstract

Humanity is facing a gloomy scenario due to global warming, which is increasing at unprecedented rates. Energy generation with renewable sources and electric mobility (EM) are considered two of the main strategies to cut down emissions of greenhouse gasses. These paradigm shifts will only be possible with efficient energy storage systems such as Li-ion batteries (LIBs). However, among other factors, some raw materials used on LIB production, such as cobalt and lithium, have geopolitical and environmental issues. Thus, in a context of a circular economy, the reuse of LIBs from EM for other applications (i.e., second-life batteries, SLBs) could be a way to overcome this problem, considering that they reach their end of life (EoL) when they get to a state of health (SOH) of 70–80% and still have energy storage capabilities that could last several years. The aim of this chapter is to make a review of the estimation methods employed in the diagnosis of LIB, such as SOH and remaining useful life (RUL). The correct characterization of these variables is crucial for the reassembly of SLBs and to extend the LIBs operational lifetime.

Keywords: second-life batteries, RUL/SOH estimation, circular economy, energy storage, Li-ion battery

1. Introduction

The Sustainable Development Goals (SDGs) are a call to action against global issues in the twenty-first century [1] such as climate change, geopolitical topics, overgrowing population, increasing energy demand, and resource scarcity, among others [2]. According to the International Energy Agency (IEA) statistics, the electricity and heat producers and transport sector are the largest greenhouse gas emitters, with at least 90% of the total CO₂ emissions [3, 4]. In 2018, above 26% of electric energy was generated from renewable sources (RSs) [5]. However, this percentage is still low in order to maintain global warming below the 2°C increment threshold stated in the 2015 Paris Agreement [6]. Taking this into account, its clear

that humanity must implement disruptive strategies to tackle these challenges. In this regard, electricity generation with RSs and electric mobility (EM) have become two of the main mechanisms for the decarbonization of the power and mobility sectors. In this context, electrochemical energy storage (EES) is a fundamental technology to realize these energy transitions by coupling both sectors in this time in history and transforming RSs from an alternative to a reliable source.

The most familiar EES devices are batteries. Compared to other energy storage mechanisms, the energy capacity of batteries is relatively low, but its efficiency is high (>95%) [7]. This makes batteries an ideal energy storage system for small- and large-scale applications [8]. According to Garcia-Tamayo [9], the convenience of batteries lies in the wide range of sizes in which they may be manufactured or assembled into packs, their ability to supply electrical power instantly, their portability (for smaller sizes), and the option of single-use or multiple-use units. The World Economic Forum reported that batteries could enable 30% of the required CO₂ reductions in the transport and power sectors, provide access to electricity for 600 million people who currently lacking access, and create 10 million safe and sustainable jobs around the world [10]. Also, since the use of internal combustion engine (ICE) vehicles accounts for a large portion of the daily energy consumption, a continuous increase of batteries through electric vehicle (EV) adoption might lead to improve grid stabilization.

Li-ion batteries (LIBs) are the most common batteries available at present and are found in almost all commercial EVs today. The battery packs inside a vehicle are composed of modules connected in series or parallel to reach the energy output and power required. Each module, on its turn, is also composed of Li-ion cells connected in series or parallel. Thus, a Li-ion cell acts as a fundamental brick of today's battery systems. A schematic illustration can be found in **Figure 1**. When an electrical load (i.e., electric vehicle, solar panel/electrical grid) is plugged and the circuit closed, during discharge, electrons (green circles) flow from the anode to cathode creating an electronic current. Likewise, Li-ions (yellow circles) are flowing in the same direction (from anode to cathode), thus converting chemical energy into electrical energy. Ions move between the electrodes by means of an electrolyte which has the property to be a good electronic insulator and good ionic conductor. As a liquid electrolyte is used in most of the cases, a separator is placed in the middle in order to maintain an even spacing between both electrodes. This separator must provide blocking of electronic current and permeation of its ionic analogue. The process shown in the schematic occurs during cell discharge. During charge, an external voltage is applied to the circuit, forcing electrons and

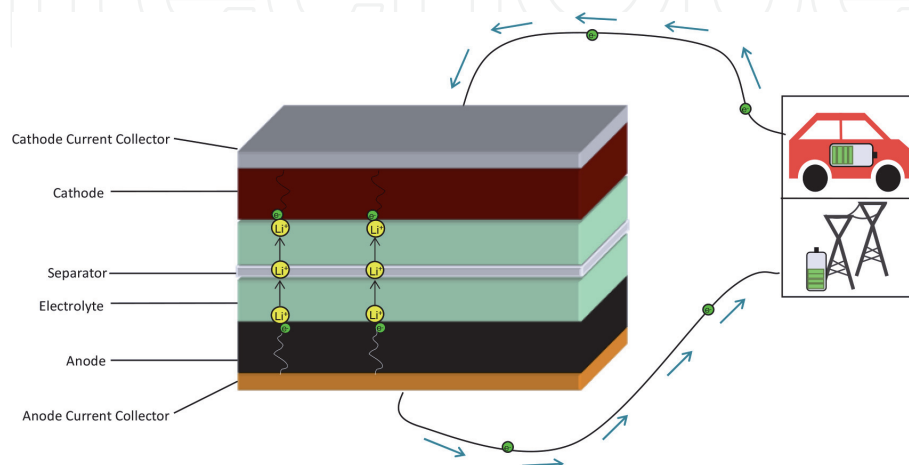


Figure 1.
Schematic of a Li-ion cell during discharge.

ions to flow from the cathode to anode. This process is performed to convert electrical energy back to chemical energy.

In general, commercial LIBs have highly pure graphite as active material for anode and different transition metal oxide lithium compounds as active material for cathode, such as $\text{LiNi}_{0.33}\text{Mn}_{0.33}\text{Co}_{0.33}\text{O}_2$ (NMC 111), LiFePO_4 (LFP), and LiCoO_2 (LCO), among others. All these cathode materials are found in commercial batteries and are referred to in the literature as battery or cathode chemistries. However, it is important to clarify that all of them are LIB technologies.

Despite the positive attributes previously described for LIB systems, there are also a set of critical characteristics that affect the battery behavior with time and as a result of their usage. The sum of these effects is a process commonly referred in literature as battery degradation or aging, which affects the cells' ability to store energy and meet power demands and, ultimately, leads to their end of life (EoL). LIBs are sensitive to the way they are charged and discharged, especially in extreme conditions such as overcharge and deep discharge as they increase the aging effect. Thus, it is of utmost importance for any device powered by LIBs to be informed of the amount of energy that can be stored and the power that can be provided by the battery at any point in time. However, the rates at which these variables degrade over time cannot be directly measured in real applications, so they must be inferred indirectly using methods and models that use input data that can be measured during charge or discharge operations.

Degradation in Li-ion cells is caused by a large number of physical and chemical mechanisms, such as active material particle cracking during Li-ion insertion and de-insertion, formation of a passivating layer on the anode/electrolyte interphase during the first cycles (solid electrolyte interphase, SEI), SEI decomposition and precipitation in the electrolyte, lithium plating and dendrite formation that could cause internal short circuit, and dissolution of transition metals from the cathode in the electrolyte, among multiple others. Multiple reviews can be found in the literature summarizing and describing in detail these aging mechanisms [11–13].

Fabrication of LIBs uses key and critical raw materials, whose exploitation and market are associated to unequal distribution of the mineral resources in the world [14]. Although lithium is a key ingredient in LIBs, manufacturers commonly use lithium carbonate or lithium hydroxide in batteries rather than pure metallic lithium. They also include other metals, such as cobalt, graphite, manganese, and nickel. Among them, cobalt and lithium are the most constrained materials [15], and nickel is important in recycling and is highly toxic to the environment. According to the US Geological Survey, worldwide lithium supply had an increase of around 23% from 2017 to 2018, coming in at 85,000 metric tons (MT) of lithium content [16]. Harper et al. estimated that the 1 million EVs that were sold in 2017 together account for nearly 250,000 MT of batteries [17]. BloombergNEF recently reported that 2 million EVs were sold in 2018, from just a few thousand in 2010, and there is no sign of slowing down. Annual passenger EV sales are forecasted to rise to 10 million in 2025, 28 million in 2030, and 56 million by 2040 [18].

In a rough approximation, if a full electric vehicle with a 33-kWh battery pack requires ≈ 5.3 kg of Li, just the EVs sold in 2018 may have required $\approx 10,600$ MT of lithium content. If battery capacities will have an increase of at least 1.8 times by 2025 (i.e., in 6 years the capacity for the Ford Focus EV raised from 23 kWh in 2012 to 33.5 kWh in 2018, while the Renault Zoe changed from 22 kWh in 2012 to 51 kWh in 2019), the EV market will require $\approx 93,000$ MT of lithium content (assuming the design or battery chemistries will not change over time), exceeding current world production. Therefore, there is still not a clear way to use less metal without compromising life span or energy storage capacity.

At present, EV batteries, most of them based on Li-ion technology, have a useful lifetime (defined by the loss of capacity due to degradation until they reach 80% of their nominal capacity) of around 300–15,000 cycles, depending on the conditions in which the battery is charged and discharged [19]. However, it is likely that they will be changed before they reach the 80% threshold not because they do not work properly but because there are other battery technologies and chemistries that will get better in the near future. For example, a recent study by Professor Jeff Dahn's group at Dalhousie University and Tesla Canada presented a LIB testing benchmark where they included a battery with a lifetime of around 4600 cycles (1.600,000 km driving range), at extreme discharging conditions (i.e., bringing the battery to a full discharge in each cycle), which could also be employed in energy storage for 20 years after reaching its EV end of life [20]. Still, even if novel batteries will get more cost-effective and safer, the battery manufacturing processes remain energy-intensive [21].

When EV batteries reach their end of life, i.e., when they reach the 80% threshold, they can still store enough energy and can operate perfectly in other uses, opening the possibility to extend their operational lifetime into a second one. Such use has been recently termed as second-life batteries (SLBs). SLB management and their possible applications are receiving a lot of attention because they could serve as a tool against the issue of 'waste' batteries being stored before repurposing or final disposal and could also save many tasks related with the managing, chemical and mechanical dismantling, and separation processes that recycling entails. To put it in perspective, the future 10 million EVs that will be sold in 2025 [18] account for nearly $\approx 2,200,000$ MT of batteries [22], which, in the absence of a second life, would otherwise end up as waste. Moreover, in the waste management hierarchy, reuse is considered preferable to recycling [17].

According to the Advanced Battery Consortium (USABC), and in most literature related to electric mobility [23], the end of life for an EV battery is defined as a 20% drop of cell capacity from the nominal value or a 20% drop from the rated power density at 80% depth of discharge (DoD, defined as the fraction or percentage of the capacity which has been removed from the fully charged battery). Nonetheless, among other factors, from an electrical and electrochemical standpoint, in order to classify the delivery of SLBs as a capable and efficient energy storage system, its remaining capacity, power, and functionality must be properly identified.

A circular economy framework diagram for LIBs is shown in **Figure 2**: (i) Used batteries from EVs that have reached their end of first life are collected. Usually their state of health (SOH) is unknown but should be around 80%. (ii) SOH testing of the battery pack/module/cell is needed to characterize its remaining capacity as compared to its initial capacity. (iii.a) The battery is depleted if the SOH is less than 40%, (iii.b) It is still usable if SOH is greater than 40%. (iv) The battery is sent for repurposing. If needed it might be broken down into its fundamental parts (cells) to connect it in series or parallel to obtain the desired energy output power for each specific application. (v) At this point, the repurposed system becomes a second-life battery and is placed on the market as a new product to serve in a second-life application. (vi) The SLB is collected after reaching its end of second life, and step (ii) is repeated to check if a third-life application is possible. (vii) If not, the battery is sent for recycling where the raw materials will be recovered and restored. Finally, the recovered materials are sent for the remanufacture of new products such as the production of new Li-ion batteries (where the whole cycle would start over).

It is important to remark that step (ii), i.e., SOH testing, is crucial to determine if the battery is depleted and immediately goes to recycling or if it may be used as a SLB for other applications. In this chapter, we will review the diagnostic and prognostic methods needed to estimate the battery current storage capacity, the

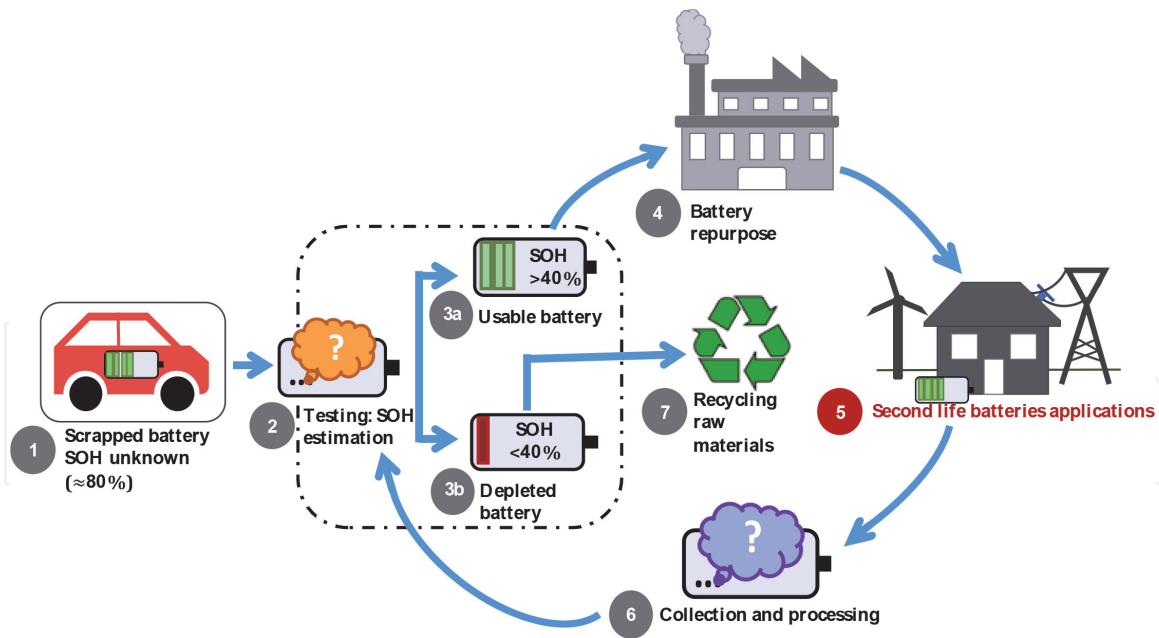


Figure 2.
 Li-ion battery circular economy framework diagram.

state of health, and the remaining useful life (RUL), which are key variables that will provide the inputs needed to define possibilities for SLB applications.

2. Review methodology

A systematic review methodology was employed as a screening method to select the information. Scopus was used as scientific database, using the following keywords as query string: Li-ion-batteries AND soh OR rul AND estimation methods AND electrochemical model OR second-life batteries. These keywords were chosen to narrow the scope of this review chapter to those focusing only on estimation methods that could be extended from SOH percentages below the 70–80% electric mobility threshold to scenarios for stationary energy storage applications that use SOH percentages that can go as low as 40%. This screening method resulted in 152 articles. A further selection was done after analyzing the title, abstract, keywords, and paper content. We identified and analyzed 15 papers which included journals and conference proceedings. The selected 15 references were studied in detail to extract useful information such as type of estimation method, estimated variables (SOH/RUL), experimental conditions, minimum SOH reached, and reported error.

3. Estimation methods

Before reviewing and establishing a classification of the estimation methods, it is important to provide definitions of the main variables found in the literature.

State of health is a percentage that measures the remaining capacity of an aged battery compared to the capacity when it was fresh. It is defined by Eq. (1).

$$SOH = \frac{Q_{actual}}{Q_{nominal}} \times 100\% \quad (1)$$

where Q_{actual} and $Q_{nominal}$ represent the actual capacity and the nominal capacity, respectively.

Remaining useful life is an estimation of the remaining time or number of cycles until the SOH of a battery reaches a specific threshold usually defined by an application. For example, in electric mobility, it is calculated until the SOH reaches 80%. Although in the literature some authors define the RUL as the time in which the SOH of the batteries reaches 0% [24], there are few articles in which the SOH is estimated below the 80% threshold.

One of the main aspects for RUL estimation is to have an accurate knowledge of the current battery state of health [25]. In the case of RUL for SLBs, it is crucial to know the minimum SOH requirements for each application in order to estimate the number of cycles or the remaining time that the batteries will last [26, 27].

In general, estimation methods for SOH and RUL are described separately in the literature [28–30]. Some authors have classified battery models for SOH diagnosis as *electrochemical*, *electrical*, and *mathematical models* [31], while others have grouped them as *direct measurements*, *model-based*, and *adaptive techniques* [32]. Similar categorizations can be found in the literature for RUL estimation methods and have been organized as *adaptive filter*, *intelligent*, and *stochastic techniques* [28]. Particularly, the classifications made by Saidani et al. [33] and Liao et al. [34] are interesting as they introduce a comprehensible way to group both SOH and RUL estimation methods in three categories, based on system theory concepts: *white-box*, *black-box*, and *gray-box* methods (see **Figure 3**). In general, these concepts refer to the level of theoretical or experimental knowledge needed to describe or model a process. Each set will be discussed in detail, but in summary white-box methods try to elucidate what happens inside a battery in terms of aging and degradation, while black-box methods employ mathematical and stochastic equations to establish correlations between intrinsic electrochemical mechanisms and external variables that can be easily measured. Gray-box methods are hybrid prognostics between white- and black-box methods where both internal mechanisms of batteries and data-driven models are integrated.

3.1 White-box methods

White-box models refer to methods that consider internal reactions and aging mechanisms of the batteries, which include physicochemical, electrochemical, and thermodynamic theories [35]. For instance, Fu et al. [36] developed a degradation model based on partial differential equations (PDEs) that estimate the capacity

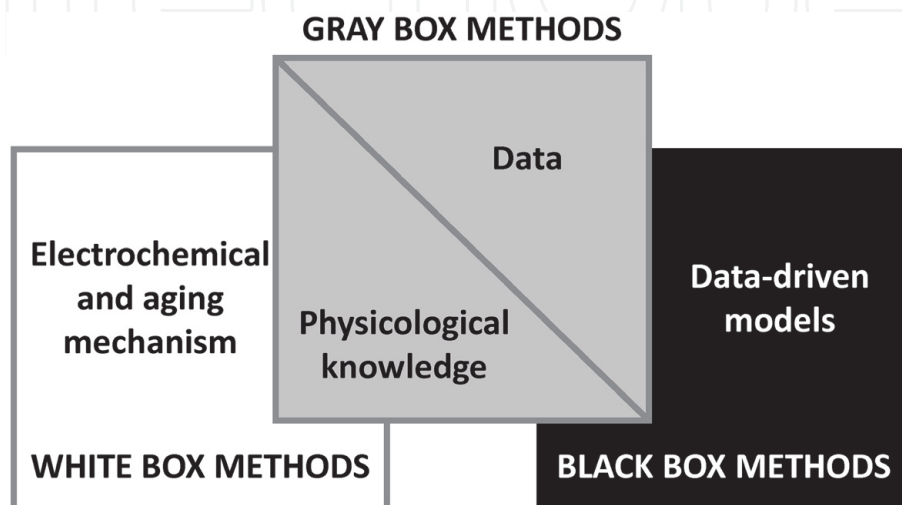


Figure 3.
Classification of SOH and RUL estimation methods.

fade using three key parameters: (i) the volume fraction of accessible material in the anode, (ii) ionic and electronic resistance of the solid electrolyte interphase and deposited layers on the electrode surfaces, and (iii) diffusion coefficient of the electrolyte. These parameters must be estimated through experimental tests and validated by characterization techniques such as scanning electron microscopy, X-ray diffraction, or X-ray photoelectron spectroscopy for each battery chemistry. This model exemplifies two of the main disadvantages of white-box methods: the need to estimate a lot of parameters and the solution of complex PDE systems. Most of the times, white-box methods derive results that are not cost-effective [33, 37].

Similarly, Gao et al. [38] proposed an electrochemical aging model that estimates the capacity fade considering the change of the open-circuit voltage (OCV) over the life span of a Li-ion battery. They reported a maximum error of about 2% for different batteries charged and discharged at different current rates (C-rates), namely, 1C, 2C, and 3C. However, this error tends to increase at the final phase of the cycling test. Likewise, with the purpose of reducing the complexity of electrochemical models, there are other methods such as single-particle models (SPMs), which assume each electrode as a single particle in order to obtain an ordinary differential equation system that models the Li-ion battery behavior [39–41]. SPMs have been integrated with a capacity degradation model coupled to a chemical/mechanical degradation mechanism that allows the prediction of the capacity fade as a function of battery temperature and cycling. The root mean squared errors (RMSEs) in these estimation methods were 7.21×10^{-3} , 7.43×10^{-3} , and 10.3×10^{-3} for LiFePO₄ (cathode)/graphite (anode) batteries tested at 15, 45, and 60°C, respectively [42].

On the other hand, white-box methods have not been used for RUL estimation due to the reasons mentioned above, i.e., because of the complexity of the models and the fact that cycles are not explicit on most of this type of methods. Thus, it is difficult to obtain parameters for SLBs' RUL because the information of the batteries on their fresh state is normally unknown [43]. However, some authors have used empirical approximations, such as Arrhenius equation (takes temperature as an accelerated aging factor) and power law (takes mechanical/electrical stress as an accelerated aging factor), to model capacity loss on batteries as a function of cycle number [30, 44].

As a result, the implementation of these methods on SLBs has been relegated since most of them do not consider the C-rate as an explicit parameter on their aging models. SOH and RUL estimation for SLBs should consider the load profile of each future application in terms of the current (amperes) needed [26, 45]. These methods have been developed for automotive applications where batteries reach their EoL when they get to a state of health of 70–80% [46] and where the capacity degradation is approximately linear until this SOH threshold, as shown in **Figure 4**. After this point, the aging behavior changes and nonlinearities start to appear [47, 48].

3.2 Black-box methods

Black-box methods take advantage of data-driven models that establish relationships between unknown intrinsic electrochemical mechanisms and external measurable variables of a Li-ion battery (e.g., voltage, current, temperature, capacity) [23]. These methods extract relevant aging features and construct degradation models based on mathematical and stochastic equations to estimate the SOH and thus predict the RUL [49]. Indeed, aging feature extraction is crucial to obtain

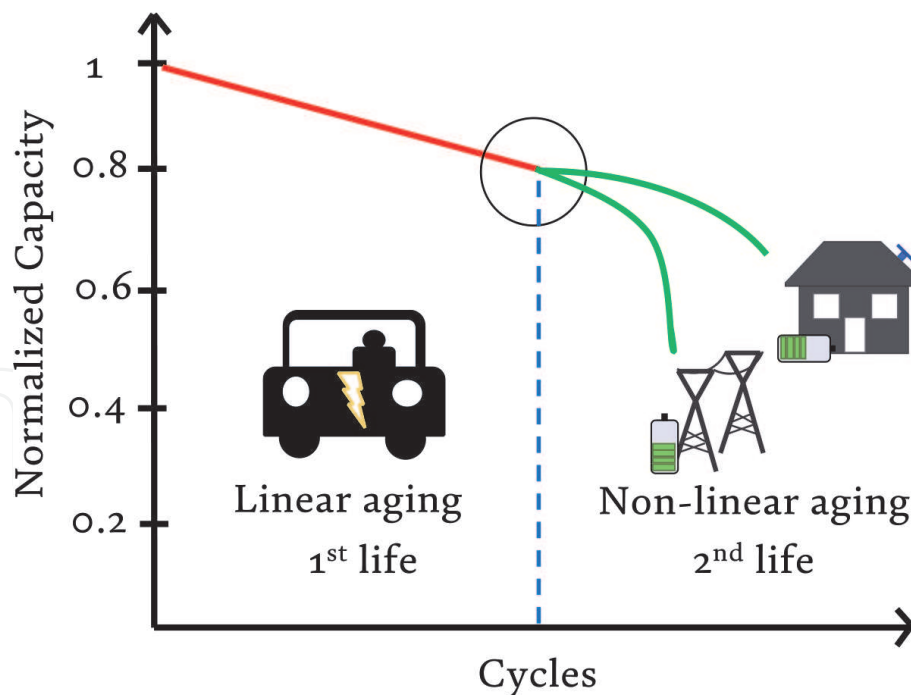


Figure 4.
Illustrative capacity degradation curve for a common Li-ion battery at its first and second life.

accuracy estimations with these kinds of methods [50]. Jiang et al. [51] tested six LiFePO_4 batteries, scrapped from a retired battery pack of an EV, with different load profiles simulating frequency regulation and peak shaving applications. They used the incremental capacity analysis (ICA) obtained from a curve of voltage (V) vs. charge/discharge capacity (Q) using Eq. (2), to develop a linear regression, constructed with the ordinary least squares (OLS) method, that could correlate features from the IC curve and the battery SOH. They obtained a mean absolute error and maximum error within 2%. Similarly, Quinard et al. [52] concluded that the ICA technique, used for SOH estimation in SLBs, has a high dependence on the C-rate (i.e., an inverse relationship between C-rate and accuracy). They reported a maximum absolute error of 5%.

$$IC = \frac{dQ}{dV} \quad (2)$$

Likewise, machine learning algorithms have been widely used in battery prognostics as these techniques can extract patterns from battery datasets, such as those from NASA [53] and University of Oxford [54], where batteries were tested at different aging conditions (C-rates and temperature). Support vector machines (SVM) [55], artificial neural networks [56, 57], and fuzzy logic [58] are some of the strategies used for SOH estimation. Nevertheless, to guarantee low-error predictions and robustness against noise, machine learning algorithms need an amount of cycling data corresponding to at least 25% of the whole battery life span [59], which could take months or years to be generated.

Taking this into account, Cai et al. [60] developed a novel method based on a combination of SVM for regression (SVR) and a genetic algorithm that employs short-term features extracted from the voltage response under a current pulse test that lasts just 18 seconds. Therefore, this process can be implemented in real SLB applications. As a result, they obtained a minor RMSE of 19.12×10^{-3} for a battery with a LiFePO_4 chemistry compared to a RMSE of 24.8×10^{-3} obtained by a traditional SVR-based model for a LiCoO_2 chemistry [61].

Another strategy that has been used to address the issues for these data-driven methods was proposed by Tang et al. [62]. They developed a model migration-based algorithm to predict the battery aging trajectory and the RUL with a notable reduction of experimental tests. This approach generates a well-known base model with enough data that is then employed in an analogous process with less available data. In this case, the base model takes advantage of accelerated aging tests, while the analogous process uses normal aging tests. As a highlight, they reached a RMSE of about 2% in RUL prediction making use of 15% of the aging data.

It is important to mention that some data-driven models extract multiple features from LIBs that do not necessarily enhance the prediction due to an emergence of redundant information [63], whereby a sliding window-based feature extraction [63] and false nearest neighbor [64] algorithms have been implemented.

3.3 Gray-box methods

Gray-box methods are hybrid prognostics between white and black methods. In other words, this category integrates both internal mechanisms of batteries and data-driven models. Liao et al. [65] stipulated that including general aging progression (white-box methods) improves the prediction accuracy of black-box methods. Equivalent circuit models (ECM) have been commonly used to simulate internal parameters such as electrochemical systems in battery management systems (BMS) [45, 66]. For instance, Wei et al. [61] modeled the capacity and impedance degradation parameters using SVR and ECM, respectively. Also, they employed particle filter (PF) to improve the SVR simulation. Tracking these aging characteristics, they estimated SOH and RUL with a high accuracy compared to an artificial neural network-based model. Likewise, references [67, 68] developed a promising modified PF algorithm that avoids particle degradation. For example, Shi et al. [68] demonstrated that improved unscented PF (IUPF) had better accuracy than unscented Kalman filter (UKF) and unscented particle filter (UPF) model prediction of ohmic internal resistance (R_o) and SOH.

In the same way, Tian et al. [69] tested three commercial $\text{LiNi}_{0.33}\text{Mn}_{0.33}\text{Co}_{0.33}\text{O}_2$ (NMC) batteries, considering the effect of temperature and discharge rate on aging cycle, to develop an on-board SOH estimation. Their model consisted in a fractional order model (FOM) using Thevenin ECM with the forgetting factor recursive expanded least square (FFRELS) method to estimate the open-circuit voltage which was then correlated to the SOH using the ICA method. Their proposed method obtained a capacity fade with an error of less than 3.1%, independent of the C-rate aging cycles.

Similarly, Guo et al. [70] used an EDKF-based model and second-order RC circuit model to estimate the SOH, obtaining a maximum error below 4%. Hu et al. [71] achieved accurate results for SOH estimation with a relative error within 3%, using a modified moving horizon estimation (mMHE) method integrated to first-order RC ECM.

3.4 Estimation method summary

A comparative summary of the SOH and RUL estimation methods mentioned above, which are included in the 15 references that resulted from the screening method described in the review methodology, can be seen in **Table 1**. For each method, it compares the employed aging feature and the reported error. Finally, there is a column for the minimum SOH reached in order to identify promising methods for SLB estimation.

Authors	Estimation method	Estimated variables	Experimental conditions*	Aging features employed for estimation	Minimum SOH reached	Reported error**
Bartlett et al. [72]	Reduced-order electrochemical model for a composite electrode battery with solid particle and liquid sub-models WHITE BOX	SOH	<ul style="list-style-type: none"> Chemistry: LMO-NMC (15 Ah) The cells were cycled using the charge-depleting (CD) current profile defined by the US Advanced Battery Consortium 	Loss of cyclable Li-ion that causes a shift of the normalized concentration operation ranges of the electrodes	≈85%	SOH estimation was performed on five different automotive cells tested at different conditions Mean estimate error: below 0.48 Ah
Li et al. [42]	Single particle-based degradation model WHITE BOX	SOH	<ul style="list-style-type: none"> Chemistry: LFP (2.2 Ah) Conditions shown in [73, 74] 	<ul style="list-style-type: none"> Cycle number Temperature 	≈76%	Error for predicted battery capacity fade RMSE: 10.3×10^{-3}
Gao et al. [38]	Order-reduced electrochemical model considering side reactions WHITE BOX	SOH	<ul style="list-style-type: none"> Chemistry: NMC (26 Ah) Ch: 1C (CCCV) protocol Followed by a 30 min rest Dch: 1C (CC) Ambient temperature: 25°C 	Capacity fade with the help of equilibrium electrode potential	60%	For cycles at 1C, 2C, and 3C Maximum error is mostly <2%
Lin et al. [58]	Fuzzy logic identification based on the closest normal distribution BLACK BOX	SOH	<ul style="list-style-type: none"> Chemistry: LCO (3.7 V/ 2.37 Ah) Ch: 0.5 C (CCCV protocol) Dch: 0.2, 0.4, 0.6, 0.8, and 1C (CC) Temp: 0–45°C 	<ul style="list-style-type: none"> Battery charging time OCV difference between fully charged battery and with a load Voltage difference between fully discharge and after resting for 1 min 	≈70%	Average error of good diagnosis: 1.46%
Long et al. [74]	Autoregressive model and the improved particle swarm optimization algorithm BLACK BOX	RUL	<ul style="list-style-type: none"> CALCE dataset: LCO (1.1 Ah) Ch: 0.5C (CCCV protocol) Dch: 0.5C (CC) Ambient temperature 	Capacity degradation	80% (defined threshold: 211 cycles)	RUL prediction difference at: Cycle 110: 26 cycles Cycle 140: 1 cycle Cycle 150: 0 cycle Cycle 190: 1 cycle
Zhang et al. [56]	Three-layer back propagation artificial neural network model BLACK BOX	SOH	<ul style="list-style-type: none"> Batteries from Beijing Olympic EV bus 	Internal resistance	Not reported. But they reach the 80% SOH from its use on second life	<ul style="list-style-type: none"> Average absolute error 0.899 Ah Capacity estimation error within 2.5%

Authors	Estimation method	Estimated variables	Experimental conditions*	Aging features employed for estimation	Minimum SOH reached	Reported error**
			<ul style="list-style-type: none"> Internal resistance measure under (100 A, -200 A, and -300 A) pulsed current tests 			
Zhou et al. [75]	Simple linear regression BLACK BOX	SOH	<ul style="list-style-type: none"> Chemistry: LCO (1.1 Ah) Ch: 0.5C (CCCV protocol) Dch: 1C (CC) 	Integral from voltage series between 3.85 and 4.3 time on CC charging phase	≈75%	Average R ² : 0.97 Average RMSE: 0.01
Cai et al. [60]	Support vector regression and genetic algorithm BLACK BOX	SOH	<ul style="list-style-type: none"> LFP (3.3. V/2.5 Ah) Load profile of primary frequency regulation Ambient temperature: 25°C 	Keen points in the voltage response under current pulse test	≈84%	RMSE for Cell 1: 19.12×10^{-3} Cell 2: 13.14×10^{-3}
Jiang et al. [51]	Incremental capacity analysis with multiple linear regression model and OLS estimation BLACK BOX	SOH	<ul style="list-style-type: none"> LFP (60 Ah) obtained from a retired battery pack Load profiles of: <ul style="list-style-type: none"> Frequency regulation application Peak shaving application Ambient temperature: 25°C 	Evolution of normalized peaks of the incremental capacity curve	≈65%	Average errors for OLS regression: MAE (%):0.609 ME (%):1.226 RMSE: 0.589
Wu et al. [76]	Neural network model with a bat-based particle filter algorithm BLACK BOX	RUL	<ul style="list-style-type: none"> CALCE dataset: LCO (1.1 Ah) Ch: CCCV protocol Dch: 1C (CC) NASA dataset LiCoO₂ (2.1 Ah) <p>Test (1) for periods of 5 min:</p> <ul style="list-style-type: none"> Ch: Series of random current Dch: CC <p>Test (2) 2A charging/discharging test after about 5 days</p>	Cycle number or cycle time	80% (CALCE defined threshold: 602 cycles) (NAS2 defined threshold: 146.83 days)	<ul style="list-style-type: none"> Capacity degradation fit: R² > 0.98 RMSE < 0.04 RUL predictions: <ul style="list-style-type: none"> For CALCE: AE: 2 cycles (at 500 cycles) For NASA: AE: 2.19 days (at 100.02 days)
Quinard et al. [52]	Partial coulometric counter BLACK BOX	SOH	<ul style="list-style-type: none"> LMO-LNO (3.75 V/65 Ah) Full CC discharge at 1C forerun by a wake-up cycle (partial charge) 	Partial capacity from a partial charge	≈45%	For partial counter: R ² : 0.69 Average AE: 1.6

Authors	Estimation method	Estimated variables	Experimental conditions*	Aging features employed for estimation	Minimum SOH reached	Reported error**
			<ul style="list-style-type: none"> • Sampling frequency: 10 Hz • Ambient temperature: 25°C 			Maximum AE: 5.1 Estimated test time: 300 s
Casals et al. [77]	Aging model based on an equivalent electric circuit that simulates the battery's behavior GRAY BOX	SOH/RUL	Real demand area regulation profile from the Spanish operator "Red Eléctrica" given to a gas turbine power plant	Current (load profile) and temperature	Considering two SLB applications on providing area regulation service: Application 1: ≈51% Application 2: ≈46%	Considering two SLB applications on providing area regulation service: Application 1: deviation of 7.35% Application 2: deviation of 8.1%
Wei et al. [61]	Support vector regression-based state-space model, equivalent circuit, and particle filter GRAY BOX	RUL/SOH	<ul style="list-style-type: none"> • Gen 218,650-size LIBs • Ch CCCV: 1.5 A CC until 4.2 V and CV continue until 20 mA • Dch CCCV: 2 A CC until 2.7 	Aging features extracted from CV protocol	≈65%	RMSE SOH SVR-PF [mΩ] #5 (5.1) #6 (8.7) #7(6.6) #18 (5.7) RUL prediction difference below 4 cycles
Tian et al. [69]	Online OCV estimation based on FOM and FFRELS GRAY BOX	SOH	Commercial NMC T: 10, 25, and 40°C <ul style="list-style-type: none"> • Ch: 1C • Dch: 1C, 2C, and 3C 	ICA peaks	60%	Capacity fade error less than 3.1%
Hu et al. [71]	mMHE integrated to first-order RC ECM GRAY BOX	SOH	Panasonic NCR18650B (3.35 Ah) at 25°C with maximum voltage and current 5 V and 100 A, respectively	ECM parameters	Not reported	Relative error of capacity within 3%

*Conditions: Ch: charge conditions; Dch: discharge conditions; CCCV: constant current-constant voltage charging protocol.

**Errors: AE: absolute error; MAE: mean absolute error; ME: maximum error; RMSE: root mean squared error.

Table 1.
Comparative summary of SOH and RUL estimation methods.

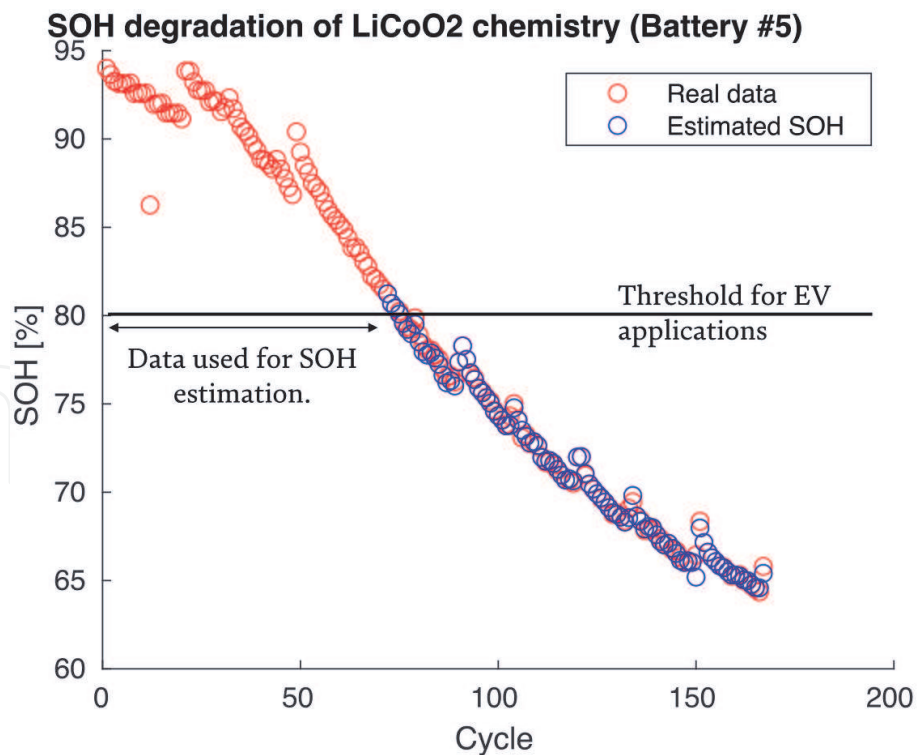


Figure 5. SOH estimation results for battery #5 from NASA dataset [53]. Model constructed using data before battery reach the 80% of its nominal capacity (authors' results).

3.5 Brief discussion on the adaptability of EV estimation methods to SLBs estimation methods

As it has been discussed throughout this chapter, there is a lack of literature for SOH and RUL estimation methods validated for SLBs. In contrast, SOH and RUL variables have been extensively studied for first-life applications for EVs. Although some published works have developed approaches for diagnosis and prognostics of SLBs applied to real second-life scenarios, such as [26, 51, 52, 56, 77], we wanted to check if a SOH estimation method developed for EV application, designed for a SOH value of 80%, could be extended to SOH values below this threshold. Hence, the black-box method proposed by Zhou et al. [75] was used for this purpose. This method calculates the integral under the constant current section of a current-voltage curve, which was obtained using the constant current-constant voltage (CCCV) charging protocol as an aging feature. **Figure 5** shows the SOH estimation for a battery with LiCoO₂ chemistry until SOH values as low as 65%. An RMSE of 0.2140 was obtained. Therefore, the authors believe that SOH and RUL estimation methods commonly employed for electric vehicle applications could be extended to estimate these variables in SLBs. However, to guarantee a better accuracy, different battery degradation behaviors must be considered depending on the load profile for each future use.

4. Conclusions and final remarks

Electrochemical energy storage in the form of Li-ion batteries is proving to be a fundamental technology to catalyze an energy transition towards renewables and electric mobility. The EV worldwide fleet, and thus the amount of batteries, is expected to grow considerably in the following years. When EV batteries reach

their end of life (SOH \approx 80%), they can still store enough energy and can be used in other applications as second-life batteries. Otherwise, they would end up as waste. It is in this context, under a circular economy scenario, that retired EVs are regarded as a primary source of SLBs. In order to do this, an accurate estimation of the state of health and remaining useful life is crucial to determine if the battery is depleted and goes to recycling or if it may be used as a SLB. Thus, sophisticated SOH and RUL estimation methods are needed to guarantee the correct performance of SLBs in different applications.

In this review chapter, we classified these methods in three categories, namely, white-box, black-box, and gray-box, which refer to the level of theoretical or experimental knowledge needed to describe the aging process in batteries. Each category has its advantages and disadvantages, and its implementation will ultimately depend on the context it will be applied. White-box methods, which are usually employed in laboratory environments, are important because they elucidate what happens inside a battery in terms of aging/degradation, and, usually, the estimation errors are lower. However, they imply the use of complex physicochemical and mathematical models and require a higher computational cost. Black-box methods, commonly employed in commercial battery management systems, make use of mathematical and stochastic equations to establish correlations between intrinsic electrochemical mechanisms and external variables that can be easily measured. Although their computational cost is usually low, they need a high amount of data to establish these correlations. Finally, gray-box methods, which are hybrid prognostics between white- and black-box methods, are considered as a promising alternative for more accurate SOH/RUL estimation as they take into account both internal mechanisms of batteries and data-driven models.

In conclusion, although there is a lack of literature for SOH and RUL estimation methods for SLBs, extensive diagnostic and prognostic approaches have been developed for EV applications. The authors believe that some of these methods could be extended to estimate these variables in SLBs. However, to guarantee a better accuracy, different battery degradation behaviors must be considered depending on the energy loads of each future use. Nevertheless, batteries intended to be repurposed in second-life applications will have to compete, at the end of their first life, with improved battery technologies and chemistries that will be likely produced at lower costs in the near future.

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