



Review

A review on lithium-ion battery ageing mechanisms and estimations for automotive applications



Anthony Barré^{a,b,*}, Benjamin Deguilhem^b, Sébastien Grolleau^b, Mathias Gérard^b, Frédéric Suard^a, Delphine Riu^c

^a Commissariat à l'Energie Atomique et aux Energies Alternatives (CEA), LIST, Information, Models and Machine Learning, 91191 Gif sur Yvette CEDEX, France

^b Commissariat à l'Energie Atomique et aux Energies Alternatives (CEA), LITEN, 17 rue des martyrs, 38054 Grenoble CEDEX 9, France

^c G2Elab, UMR 5269, 38402 Saint Martin d'Hères, France

H I G H L I G H T S

- A review of the progress made for understanding battery ageing phenomena is provided.
- The ageing battery estimation methods are summarized and compared.
- The challenges and unresolved issues for battery ageing estimation are discussed.

A R T I C L E I N F O

Article history:

Received 13 July 2012

Received in revised form

4 May 2013

Accepted 10 May 2013

Available online 7 June 2013

Keywords:

Ageing

Lithium-ion battery

Estimation

State of health

Remaining useful life

Modeling

A B S T R A C T

Lithium-ion batteries have become the focus of research interest, thanks to their numerous benefits for vehicle applications. One main limitation of these technologies resides in the battery ageing. The effects of battery ageing limit its performance and occur throughout their whole life, whether the battery is used or not, which is a major drawback on real usage. Furthermore, degradations take place in every condition, but in different proportions as usage and external conditions interact to provoke degradations. The ageing phenomena are highly complicated to characterize due to the factors cross-dependence. This paper reviews various aspects of recent research and developments, from different fields, on lithium-ion battery ageing mechanisms and estimations. In this paper is presented a summary of techniques, models and algorithms used for battery ageing estimation (SOH, RUL), going from a detailed electrochemical approach to statistical methods based on data. In order to present the accuracy of currently used methods, their respective characteristics are discussed. Remaining challenges are deeply detailed, along with a discussion about the ideal method resulting from existing methods.

© 2013 Elsevier B.V. All rights reserved.

1. Introduction

Lithium-ion batteries have been commercialized since 1991, initially concerning mobile devices such as cell phones and laptops [1]. Interest on this technology has considerably increased and generated a lot of research in order to improve the performances of those batteries [2]. Recently, lithium-ion batteries penetrated the market of hybrid and electrical vehicles as a result of the high lithium's density, the weak weight of the lithium batteries making them the most promising candidate for this field of application [3].

Different organizations converged to estimate Electric Vehicles (EV) representing ~60% of the total market of passenger cars by

2050 [4,5], with a presence on all major regions of the world. Supposed evolution of EV sales, highly sensitive to the battery development, is the result of petroleum prices increasing [6]. As an example, Renault demonstrates the profitability of its Fluence ZE starting from 15,000 km year⁻¹ [7]. Considering prices evolution, this benefit is predicted to decrease with time, which will induce more interesting EV costs [8].

Such market evolution implies important coming steps for batteries as roadmaps consider the long term goals of battery evolution to be: augmentation of maximal capacity, acquiring a battery lifetime equivalent to the car's life cycle, reducing costs in order to be the same as those of an Internal Combustion Engine (ICE) vehicle, operating in all climates [9–11].

The first use of this battery technology had a low lifetime need. With the new applications, interests are now focused on ageing phenomena considering manufacturers requirements. In terms of

* Corresponding author.

E-mail addresses: anthony.barre@cea.fr, anthony.barre@live.fr (A. Barré).

battery design, beyond all performance constraints, some objectives are clearly defined for service life (10–15 years or 20,000–30,000 discharges [4]). Therefore, battery ageing phenomena are commonly used to evoke both main consequences of time and use on a battery. The resistance growth and the capacity fade, will be discussed in the following parts. The aims of increasing battery performances stoke the requirement of a better understood battery ageing [12,13].

Identifying ageing and degradation mechanisms in a battery is the main and most challenging goal. Such processes are complicated as many factors from environment or from utilization mode interact to generate different ageing effects. Hence, the capacity fade and the resistance growth do not depend on the same variables. This makes the ageing comprehension a difficult task, and throughout the years, many studies tried to explore the battery ageing.

This review intends to summarize today's results on mechanisms, factors and estimation methods of lithium-ion battery ageing on automotive applications. The first aspect presented here is the notion of battery ageing in an electrochemical description, with explanations of known battery ageing phenomena. Based on these ageing characteristics, many studies investigated lithium-ion battery ageing factors, effects, and tried to estimate the battery state of health (SOH) by several methods. These ageing studies and methodologies come from many various fields such as electrochemical models, performance models or statistical methods. The diversity and the multitude of existing studies dealing with battery ageing provide a large amount of information. This paper presents all of these approaches along with their respective performances. Finally, a discussion on the methods advantages and drawbacks is proposed. Finally we will suggest a new methodology for battery ageing estimation solving the drawbacks of the previously seen methods.

2. Electrochemical ageing

Ageing initially takes place in the chemical composition of the battery's electrolyte. The degradation mechanisms from the positive and negative electrode are different [14,15]. The origin of ageing mechanisms can be either chemical or mechanical and are strongly dependent on electrodes composition. Throughout time ageing provokes cell components degradation [16], which can induce, for instance, a modification of the structural properties, a variation of the electrolyte chemical composition, or a loss of active material by the dissolution of material in the electrolyte, such as manganese [14]. Thus, the main ageing phenomena come from degradation of electrodes.

2.1. Ageing effects on negative electrode

Most negative electrodes are composed of graphite, carbon, titanate or silicone [17]. The choice of the graphite material is important in ageing and safety properties of a battery [15]. The main ageing factor on graphite electrode with time, is the development of a solid interface on the electrolyte/electrode interface. This is named Solid Electrolyte Interphase (SEI) [18]. This solid interphase is naturally created during the first charge. Its role is to protect the negative electrode from possible corrosions and the electrolyte from reductions [19]. This phenomenon predominately occurs during the beginning of a cycle. Its a natural barrier between the negative electrode and the electrolyte and consequently provides a guarantee of security [20,21]. The SEI is not stable as lithium-ion battery operates in tension outside the electrochemical stability range of the electrolyte [22]. Thus, the SEI develops over time which induces loss of continuous lithium ions and an electrolyte decomposition [23]. Moreover, loss of available lithium due to side reactions at the graphite negative electrode has been

reported as the main source of ageing during storage periods [24]. That is, the SEI is relatively stable over time, inside the stability window, and the capacity loss is not significant in the short term, allowing lithium-ion batteries utilization over long periods.

Furthermore, the SEI is permeable to the lithium ions and to other charged elements (anion, electrons) or neutral elements (solvent) [14,25]. Thereby, the solvent interacts with the graphite after diffusion through the SEI, which induces graphite exfoliation [26] and creates gas which can crack the SEI and therefore allow its expansion [19,27]. Nevertheless, the gas formation is low and it seems to happen only during storage periods and with high voltage [28]. With time, there is a loss of active surface, increasing electrode's impedance. Fig. 1 illustrates all these phenomena occurring at the SEI [14]. This phenomenon may take place during utilization of the battery as well as during storage.

A high SOC (State Of Charge >80%) should provoke an acceleration of these phenomena as the potential difference between electrode interfaces and electrolyte is important [29]. Moreover, inadequate conditions can accelerate the process, such as high temperature, overcharge, short circuit [30]. Thus, under high temperatures, the SEI may dissolve and create lithium salts less permeable to the lithium ions therefore increasing the negative electrode impedance [31]. On the contrary, low temperatures lead to a decrease of the diffusion of lithium within the SEI and graphite [32,33], which can overlay the electrode with lithium plating. It is important to note that the SEI formation, its development, and the lithium plating are all responsible for the loss of cyclable lithium, under conditions of transportation utilization [34].

2.2. Ageing effects on positive electrode

Bourlot et al. [28] shows from positive electrode observations, that there is no evident modification of the positive electrode's morphology, for all levels of battery utilization [35]. This is the

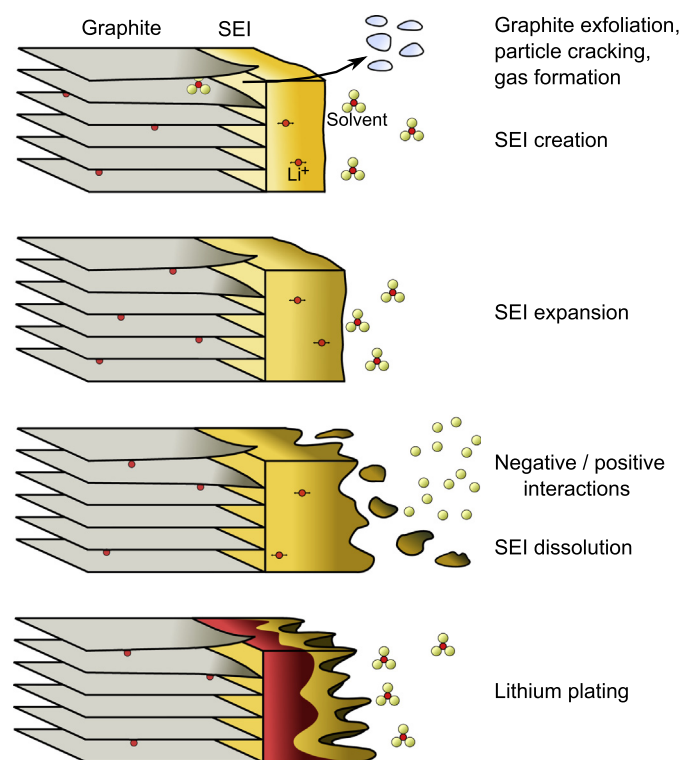


Fig. 1. Illustration of ageing effects on battery negative electrode: the capacity fade and the SEI raise.

confirmation of the primordial importance of the negative electrode in the battery ageing [36].

However, the positive electrode is subject to a low alteration within time, depending on the chosen material [37]. There is also a SEI creation on the positive electrode/electrolyte interface, that is more difficult to detect [38,39], due to high voltages on this electrode [40]. To sum up, the principal consequences observed on an aged positive electrode are: wear of active mass, electrolyte degradation, electrolyte oxidation and formation of a SEI, interaction between positive electrode element dissolved within the electrolyte and the negative electrode [35,41,42].

These effects are not independent and their respective interaction differs according to the used positive electrode material [43]. As for the negative one, statements highly depend on the SOC and the temperature.

2.3. Consequences of ageing phenomena

In this part, two principal effects of battery ageing are identified: capacity fade and impedance raise. Both phenomena differ by chemical causes, thus have different origins. This implies a non-linear dependence of these ageing impacts.

The performance loss is caused by various physical-based mechanisms, which depend on the electrode materials. They can either be of mechanical or chemical origin. The consequences of these mechanisms on the lithium-ion cells are:

- The (primary) loss of cyclable lithium which increases cell imbalance. Loss of cyclable lithium is related to side reactions. This can occur at both electrodes, as the SEI grows at carbon anode due to electrolyte decomposition [44].
- The (secondary) loss of electrode active materials, possibly a material dissolution, structural degradation, particle isolation, and electrode delamination [45].
- Resistance increase of a cell. This can occur due to passive films at the active particle surface as well as loss of electrical contact within the porous electrode [46].

In terms of battery performances, both loss of cyclable and loss of active materials lead to the battery capacity fade. Secondly, the battery resistance growth is engendered by the passive films. On vehicle utilization, the capacity loss induces an autonomy reduction. On the other hand the resistance augmentation reduces the maximum of power available.

3. Ageing origins

Battery ageing can be dissociated into two parts: the calendar ageing and the cycle one [47]. Each term defines the alterations caused by different uses of the battery. Thus, the calendar ageing corresponds to the phenomena and the consequences of battery storages. On the contrary, cycle ageing is associated with the impact of battery utilization periods named cycles (charge or discharge).

3.1. Calendar ageing

Calendar ageing is the irreversible proportion of lost capacity during storage. In other terms, it is the degradation caused by the battery storage [48,49]. Self discharge rate varies highly according to storage conditions. Hence, effects occurring within the battery can be accelerated or slowed depending on the storage conditions [50]. Numerous experimental studies showed the impact of storage conditions on this ageing. For example, studies tested cells over several temperatures, SOC (sixty cells for Bloom et al. [51] and three

hundred for Wright et al. [52]) and Ramasamy et al. [53] studied cells under different end-of-charge voltages and temperatures.

The main condition considering the calendar ageing and the self discharge is the storage temperature [54]. When the temperature is high, secondary reactions such as corrosion are facilitated and the lithium loss is more important than in moderate temperature conditions, which induces capacity fade [51,52,55]. Low temperatures enable to limit the development of these phenomena but these conditions engender some problems due to the loss of material diffusion and alter the battery chemistry [56].

The other principal variable of calendar studies is the SOC level during storage [57]. Thus, for an equal temperature but for different SOC, cells do not age in the same manner. This illustrates a higher battery degradation for elevated SOC [36]. By definition, the SOC represents the ions proportion present on electrodes. This implies, for high SOC, a huge potential disequilibrium on the electrode/electrolyte interface. This promotes precedent chemical reactions.

Most calendar studies explore SOC as a combination with the temperature storage conditions. Indeed, each of these variables alter together the capacity and the resistance with a non-linear effect with time. Studies results [58,59] evocate the more restrictive effect of a high SOC than of a high temperature. Such results are just an interpretation from a few experiences and it remains to understand the complete effect of the combination of those two variables on the calendar ageing.

Temperature and SOC impact directly battery calendar ageing. Furthermore, capacity fade and resistance augmentation are not linear with time, which implies a strong interaction of ageing behavior with time.

3.2. Cycle ageing

Cycle ageing happens when the battery is either in charge or in discharge. This is a direct consequence of the level, the utilization mode, the temperature conditions and the current solicitations of the battery. Consequently many factors are involved with this kind of ageing. All factors previously described impact the calendar ageing and are also included in studies of the cycle ageing, because ageing phenomena previously cited, appear whether the battery is used or not. In most cases, a battery in use is prone to exothermic effects [60,61] and those effects can be facilitated under high temperatures and provoke battery ageing. However, it is important to take into account the effects of very low temperatures [59]. Studies report direct impact of ambient temperature but none of them deal with direct battery temperature. This notion remains misunderstood.

Apart from such variables, cycling ageing's factors are function of the battery utilization mode. A recurrent factor on literature is the Δ SOC, which represents the state of charge variation during a cycle. This is an important factor considering the amount of charge taken (resp. given) to the battery during a discharge (resp. charge) [35,62]. Bloom et al.'s experience [51] consists in testing identical lithium-ion cells with similar temperatures and initial SOC but for different Δ SOC. Results show loss of battery power as Δ SOC's value is high and this is for all other possible conditions. This was later confirmed by other experiments [63]. Such phenomena are mainly due to the positive electrode degradation and to the SEI development, engendered by high discharge, or charge.

Another variable impacting the lithium-ion battery ageing and function of the utilization mode is the charging/discharging voltage during its life. Thus, high charging voltage implies accelerated ageing phenomenon [64]. To illustrate this, Asakura et al. [65] show that the battery life halved for a 0.1 V augmentation of the charging voltage, the EOL is considered here as 70% of initial capacity. Discharge voltage influences the battery ageing through the impedance augmentation [42,66].

Finally, current peak seems to be a notion involved with ageing phenomenon. Indeed, an important current peak induces a high level of given or released energy.

3.3. Conclusion on ageing characteristics

Previously presented factors influencing battery ageing interact to generate both capacity loss, resistance augmentation and loss of available peak power [67,68]. Note that all factors are dependent on other external conditions. For transport application, temperature depends on climate, Δ SOC depends on the driving cycle... Thus, utilization mode is widely concerned by ageing studies. Depending on utilization modes, such as successive accelerations or constant velocity, the battery temperature will not evolve the same way. Peak current is not identical within road profile and conductor aggressiveness.

These characteristics make of ageing comprehension and estimation of a huge challenge due to the multiple interactions between all factors coming from utilization mode or environment [69,70].

4. Ageing estimation

To evaluate ageing, several indicators or notions are created in order to quantify the health level of the battery. The most used indicator in the literature is the State Of Health (SOH) which is generally defined by Refs. [71,72]:

$$\text{SOH}(t) = \frac{\text{nominal capacity at } t}{\text{initial capacity}} (\%)$$

Other SOH definitions can be made through the End Of Life criterion (EOL) [73] but it is still a proportion of battery capacity remaining. This indicator represents the battery capacity fade [74,75]. As the “ageing” term is not precisely defined, other indicators such as State Of Function (SOF) [73] or Remaining Useful Life (RUL) [76] are introduced. All these notions are deducted from the capacity state of the battery and therefore do not consider all parts of ageing as the resistance is omitted. However, resistance development is specially impactful on high power applications. Thus, Ecker et al. [77] define the EOL criterion as the moment where the initial inner resistance doubles.

Different methods are used to estimate these notions of battery age level. They are divided into five parts:

- Electrochemical models: detail and model the phenomena occurring into the battery
- Equivalent circuit based models: the battery is reduced as an equivalent circuit model
- Performances based models: battery ageing is modeled by physical equations
- Analytical models with empirical fitting: estimation of ageing parameters through measurements
- Statistical approach: approaches mainly based on data, without a-priori knowledge

4.1. Electrochemical models

Battery ageing can be determined from physical models. Such models seek to quantify factors impact and therefore obtain the description of battery performance evolution. As said previously, several factors interact to generate battery ageing phenomena, which causes the complexity of making a reliable and precise model. The goal of these approaches is to give a sharp understanding of the specific physical and chemical phenomena occurring during battery utilizations.

Physical models can be distinguished into two separate parts. The phenomenological approaches giving a dynamic description of the cell which could be independent of the electrodes materials. The other part consists in atomistic/molecular models. They allow access to thermodynamics quantities related to electrodes structures, surfaces or electrolytes such as thermodynamic energies, activation barriers or reaction mechanisms.

Degradation mechanisms related to the material properties can be deduced from both these approaches. On one side the impact of an ageing process on the cell performance can be translated into physical equations by fitting the parameters using the macroscopic observations. On the other side, it is possible to identify the prominent occurring physico-chemical processes depending on the employed materials, and evaluate with atomistic calculations, their effect on the battery [78,79].

4.1.1. Phenomenological approaches

The development of these models starts about twenty years ago, with the work of the Newman's group [80,81], seeking to estimate battery performances based on Butler–Volmer equations and porous electrodes theory [82]. The first attempt of simulation ageing process in a physical model was made by Darling and Newman [83] with the implementation of a simple solvent oxidation reaction within their model. The results were very encouraging and gave good conclusions concerning solvent choice. Based on the same approach, Christensen et al. [84] explained the capacity fade with the augmentation of resistance on the negative electrode surface and they developed a model to represent the SEI [85].

Most of the mechanisms incorporated in physical models are related to the negative electrode. These models have investigated the impact of the stress occurring during the lithium intercalation process in Lithium Manganese Oxide (LMO) batteries [86,87] and managed to predict which electric current and SOC caused the appearance of fractures in active material particles. The SEI raise and its relation with the capacity loss, were also been studied [88] with a solvent diffusion model. More recently, studies [89,90] have estimated the effective diffusion coefficient in the electrolyte with the cycle number. They were able to simulate the evolution of the electrolyte composition and the consequences on the cell performances. Safari et al. [91,92] also developed a phenomenological model estimating the performance of a commercial Lithium Iron Phosphate (LFP)/graphite cell and tried to estimate the impact of the ageing on the battery performances in terms of capacity loss and impedance raise. This study permits to evaluate that cycling is more determinant than storage on degradation. The loss of active material is less important than the loss of active lithium for these batteries under temperature conditions between 25 °C and 45 °C [93].

4.1.2. Atomistic and molecular approaches

In the approaches listed above, the intrinsic properties related to the electrode and electrolyte materials are not precisely taken into account. In order to understand at a nanoscale level the phenomena appearing during the battery ageing, a few models have been developed using atomic and molecular methods.

Density functional theory (DFT) is already used to describe the different phenomena occurring during the lithiation/delithiation of the electrodes. Depending on the materials nature, these processes can follow very different mechanisms from intercalation (in the case of LiCoO₂ batteries) to the biphasic domino-cascade process (for LiFePO₄). Theoretical methods have been developed to investigate these phase transition processes. Dalverny et al. [94] performed ab initio calculations to determine the morphology of the LiCoO₂ electrode. Moreover theoretical studies have been done to understand the phase transition process within LFP active particles

[95]. Ab initio calculations can contribute to the degradation phenomenon comprehension by estimating the energies related to solvent decomposition or to lithium salts dissolution. Tasaki et al. [96,97] have already linked the lithium salts dissolution near the negative electrode's SEI with the calendar capacity fading.

Molecular dynamic also represents a good way for the understanding degradation mechanisms. Indeed, a study [98] has investigated the decomposition of the Ethyl Carbonate (EC) solvent and the consequences on the SEI raise. This could be a key point for comprehension of ageing problems related to graphite electrodes. Moreover, SEI evolution has been examined by another recent study [30] using Kinetic Monte Carlo (KMC) method. They focus their work on the diminution of the electrode active surface related to SEI formation during cycles.

One of the greatest challenges of the physical models is to link results between atomistic approaches (DFT, molecular dynamics...) and macroscopic models. The aim is to obtain a detailed description of all the phenomena occurring at a nanoscopic scale and thus estimates directly their impact on the battery performance. This objective is reachable but the remaining problem is to associate top-down models, going from macroscopic observation to a nanoscopic description of the battery, to bottom-up models.

4.2. Equivalent circuit based models

Other methods permit the estimation of ageing based on models. The model-based methods commonly modelize battery with an equivalent circuit model and use different techniques to estimate these model parameters [99,100]. However equivalent circuits have defined definitions according to studies.

In order to estimate the battery ageing, parameters can be internal battery parameters as well as resistance ageing parameters. Parameter identification can be directly made from measurements or from more complex approaches through equivalent circuit models [101–103]. Such methods require a large and diverse data set obtained with time consuming tests. For example, the Relevance Vector Machine (RVM), a machine learning method, is used in battery ageing estimation through this parameter identification. This method is a Bayesian form representing a generalized linear expression of Support Vector Machine (SVM) introduced by Vapnik [104], currently the state-of-the-art in regression and classification algorithms [105,106]. This method deals with different parameters from large data sets and estimates and predicts the battery degradation. Saha et al. [107,108] used this method to learn a dependency model of internal battery parameters model to estimate and predict the SOH. These studies also use Particle Filter (PF) and later the Rao–Blackwellized PF (RBPF) to obtain a distribution of the RUL prediction.

4.3. Performances based models

This approach uses simple correlations between stress factors and capacity fade/impedance raise. These correlations are induced from ageing tests conducted under several conditions. These methods intend to quantify the impact of ageing factors and obtain a descriptive expression of the battery performance level over lifetime. Most times studies deal independently with both ageings: calendar and cycle [77,109]. Furthermore, ageing studies are not only focused on capacity and resistance evolutions but also on equivalent circuit parameters in both cycling [110] and calendar part [77,111].

4.3.1. Calendar ageing

In calendar ageing modeling, the main variables are time and temperature. However, some studies consider another parameter:

the storage SOC [112,113]. As seen in the Part 3.1, the SOC influence cannot be neglected in certain conditions. The calendar ageing appears to follow Arrhenius-like kinetic (1), as this law is typically used in order to consider storage temperatures [24,51,114]. Thus, capacity fade and resistance increase may be mainly caused by thermal processes which are linear with time [112].

$$t = A \exp\left(\frac{-E_a}{RT}\right), \quad (1)$$

with t a lifetime, A constant, R gas constant, T temperature and E_a the activation energy.

The storage SOC is introduced by using the Tafel equation relating the rate of an electrochemical reaction to the over-potential (2), as in a modeling study [109].

$$\Delta V = A \ln\left(\frac{i}{i_0}\right), \quad (2)$$

with ΔV the over-potential, A constant: "Tafel slope", i the current density of the electrochemical reaction and i_0 the exchange current density.

In most studies, the capacity fade due to storage was observed as linear dependent on time^{1/2} [51,109,115]. For example, Ecker et al. [77] proposed a calendar lifetime prediction model describing capacity and resistance evolutions over time as shown in (3):

$$\frac{L(T, V, t)}{L(T_0, V_0, t_0)} = 1 + B(T, V) \cdot c_a \cdot t^{1/2} \quad (3)$$

with c_a coefficient of the degradation rate (capacity fade or resistance increase) under reference conditions T_0 , t_0 and V_0 . $L(T, V, t)$ stands either for resistance or capacity estimated at time t , with a temperature T and voltage V . Moreover, the impacts of temperature and storage voltage are calculated according to an exponential dependence (4):

$$B(T, V) = c_T \frac{T - T_0}{\Delta T} \cdot c_V \frac{V - V_0}{\Delta V} \quad (4)$$

where T_0 and V_0 are reference temperature and voltage. ΔT and ΔV have been arbitrarily set to 10 °C and 0.1 V. Parameters c_T and c_V of the proposed law (4) are based on accelerated calendar ageing test data. Considering these data, a time^{1/2} dependence of the degradation rates was assumed.

Numerical coefficients values are only obtained based on accelerated calendar ageing data. According to experimental data, there is a hypothesis of a minor contribution of cycle part on ageing. Simulation results are in good agreement with the capacity fade evolution during cycling but overestimate substantially the resistance over time.

4.3.2. Cycle ageing

The cycle ageing is more complex to predict as it involves more independent variables such as temperature and current voltage. Furthermore, these variables are related to external conditions as well as battery utilization. The main factors considered are typically the temperature, the cycle number, Δ SOC, voltage. Note that most studies agreed to deal with cycle number as a main time notion. Thus, Wenzl et al. [116] evoke a simple cycle counting method, but the choice of other factors is subject to discussion.

For example, on one hand a study proposes a typical cycle number dependency model for cycle ageing [109] and another one found that each model parameter follows a cycle^{1/2} trend [117]. On the other hand, Wang et al. [118] developed a cycle life model of

LiFePO₄ cells by considering ΔSOC, temperature and C-rate. This study finds a $Ah^{0.55}$ dependence of the capacity fade, where Ah is the cumulative charge delivered by the battery over its life. According to the author, this parameter is directly proportional to ageing time but allows correlating degradation for different C-rates.

In the case of separate models, both ageing models can be added with each other to represent a general ageing expression.

4.3.3. Global performance models

Another method consists in estimating ageing directly without considering a cycle or a calendar point of view. It can be the same kind of model explained previously but by considering the ageing in a general way [119,120], but also a damage-accumulation model (fatigue approach). Damage models use a stress factors/capacity fade correlation in order to fix a relationship for capacity loss over time. The Palmgren–Miner rule model is an example of damage accumulation model [121,122].

In order to use this model, a profile-decomposition method needs to be proposed to divide the load profile into pieces compatible with the stated empirical correlations. Safari et al. [123] provide a detailed description to use this method but results were compared with a theoretical graphite/LiCoCO₂ battery model subjected to only one ageing process (SEI formation at the negative electrode).

The main drawback of these models is that they do not provide any insight into the process that contributes to capacity fade and impedance raise. As a consequence, impact of each stress factors has to be investigated independently resulting in heavy sets of experiments. Moreover evaluating battery life by extrapolating accelerated test results can lead to large errors as illustrated by Takei et al. [124].

4.4. Analytical model with empirical data fitting

An empirical method is based on data, as large as possible, from experiments in order to evaluate or predict estimator values. Such methods are used to determine model parameters as well as a direct ageing estimator. The main troubles of these methods are the lack of data and the accuracy of the measurements [125].

The most popular method is the “coulomb counting” which allows estimation of SOH by a simple integration of current over time [126]. The major inconvenient of this method is the necessity to do this counting every time under same conditions, such as external temperature for example. It requires a recalibration at regular intervals and it cannot be done in real time [127].

Fuzzy logic is also used to admit a low noisy level on the data sets [128]. Thus, based on the data a fuzzy logic fixes an input–output relationship based on expertise and can estimate an ageing parameter directly or through a model [129,130]. Salkind et al. [131] utilizes this method to estimate NiMH battery’s SOH. Furthermore, Singh et al. [132] used data from Electrochemical Impedance Spectroscopy (EIS) in order to find two entry parameters: the magnitude of impedance and the phase angles. This method predicts the number of remaining cycles available for the battery use. This last method is extremely hard to implement due to the EIS requirement. It is important to note that fuzzy logic method requirements can add significant errors by the expertise assumptions.

A method based on state observations model the ageing estimation problem with an equation system (5), with an input u (state vector) and an output y (voltage), depending of variables x .

$$\begin{cases} \hat{x} = Ax + Bu \\ y = Cx \end{cases} \quad (5)$$

The goal is consequently to adjust a model from observations in order to minimize the error between \hat{x} and y through a gain K .

This correction gain K is then fixed by an algorithm, most generally a Particle Filter (PF) [133], more precisely an Extended Kalman Filter (EKF) [74,134]. The main problem of this method comes from the possible fast divergence under unsuitable conditions, because it is necessary to preset the initial matrix [135].

Artificial Neural Networks (ANN) or Neural Networks (NN) are mainly used for battery’s SOC prediction [136] but this method can also measure the battery SOH [137,138], as it was done for lead-acid battery [139,140]. Such studies take as entry variables simple parameters: voltage, discharge current, discharged capacity, regenerative capacity and temperature. The main advantages of this method are that it requires only easily obtainable values. NN performs despite multi-dependences and affirms the easy adaptivity of the method to other battery technology [141]. As any learning mechanism, NN requires a large number of diverse data to be effective.

4.5. Statistical methods

As analytical models, statistical methods require a large data set to be effective. These methods do not need any a-priori knowledge on the ageing mechanisms and there are not any hypothesis made on factors and they do not use any chemical or physical formulation.

A simple method is the use of time series process, mostly Autoregressive Moving Average (ARMA) [142]. These studies consider the ageing level data as a chronological series and the ARMA methods can deduce the following value of this ageing level [143]. Such a method only works for one battery as each ageing will be different because it depends on the battery usage. Moreover the accuracy depends on data: it requires many full battery characterizations to obtain these data. These inconveniences make this method an unrealizable solution on board applications.

Another approach can considers the battery’s end of life criteria as a failure and model this end of life by a Weibull law [144,145]. However, this method considers all different uses and conditions as a unique way which considerably reduces the result accuracy.

The methods previously stated as NN, Fuzzy Logic and RVM can also be performed by directly estimated an ageing parameter without considering an a-priori model.

5. Discussion

5.1. Assessment of estimation methods

From measurements to statistical estimations, many different kinds of empirical methods exist to estimate the level of battery ageing, each of them has pros and cons characteristics.

Direct measurements do not need battery hypothesis as it is a direct estimation. Furthermore, the only bias introduced is the measurement incertitude. However, this method performs for all kinds of battery usage. Due to the procedure duration, it is impossible to do such measurements in real time, and to predict the ageing evolution (Part 4.4). Especially for an automotive approach, it is unrealistic to do complete battery measurements to estimate its degradation level.

Methods like electrochemical models and equivalent circuit models perform well but cannot be directly extended to other batteries (technology, design, materials). This is the first drawback of these methods as technologies are constantly improving which produces new different batteries. Moreover, these two approaches are not performant to model all degradation mechanisms occurring during the battery life, but only the ageing trend in the best cases (Parts 4.1 and 4.2). This is caused by the initial assumptions. However, electrochemical and physical models are powerful tools

to understand the different interactions between different physical phenomena and the trends about operating condition effects on ageing.

The same statements can be made on the performance models as ageing is a complex and a multi-dependent phenomenon. This method is also dependent on the battery technology. Estimating ageing under controlled situation performs well but this is rarely tested on real vehicle data because a lot of environmental variables interact (Part 4.3), which makes the problem difficult to model.

On the contrary, statistical methods are easily adjustable to different batteries, and they can perform to give an ageing diagnosis in real time. But, this kind of method requires a large amount of data to be effective. This data collection process is a complex task due to the battery lifetime in real usages (>5 years on the manufacturer guarantees). The battery lifetime implies a very long data collection time. Furthermore, in order to be efficient in all situations, statistical methods must be constructed from various experiments like different usages mode (driving style, road, climate...) (Part 4.5). To sum up, the disadvantage of this approach is clearly the data collection process.

As it is explained here, none of the actual methods are not performing enough to obtain an ageing estimation able to be included in a real electric vehicle diagnosis experiment. Table 1 represents performances of methods for battery ageing estimation based on different criteria such as their prediction capacities, their abilities to perform in real time context, or their accuracies. The different methods can be complementary, and because of the degradation phenomena complexity, it is necessary to group some of them. That is, the Table 1 clearly illustrates the actual compromise we are facing with the battery ageing estimation.

5.2. Battery ageing summary

The current EV development induces high expectation on future battery performances and longevity. This requires a good battery ageing comprehension and estimation, which is still a challenging problem.

The battery ageing is hard to identify and quantify due to the diversity and the complexity of the phenomena taking place into a battery during its whole life. The two different ageings, capacity fade and resistance augmentation, are distinct as degradations provoked during storage and utilization have different influence on the battery characteristics [47].

Many variables are involved in the ageing process, having a more or less direct role [68]. The more recurrent factors are for the calendar ageing: temperature, storage SOC and time. Cycle ageing concerns temperature, Δ SOC, cycle number, charge/discharge voltage and factors coming from the utilization mode but not clearly identified yet, peak current demand for example. These variables come from internal and external conditions and interact

Table 1
Battery ageing estimation methods performances comparison for five principal aspects.

	Adaptation	Precision	Operate without data	Real time	Prediction
Direct measurement	Excellent	Excellent	Excellent	Very poor	Very poor
Equivalent circuit model	Very poor	Fair	Good	Good	Fair
Electrochemical model	Very poor	Excellent	Fair	Fair	Fair
Performance model	Very poor	Good	Poor	Fair	Good
Analytical model	Very poor	Good	Poor	Poor	Poor
Statistical method	Fair	Good	Very Poor	Good	Good

with each other. All these interactions are the main complexity of ageing comprehension, and the result of these interactions are difficult to understand and quantify (Part 3).

Studies based on test bench misunderstand the real impact of all variables due to the controlled conditions. For example, Bögel et al. [54] try to reproduce a real EV use on a test bench, but results of this study illustrate the limits of such approaches. These simulations can lead to results such as a complete linear dependence between an ageing phenomena and a variable. Such conclusions usually allow these studies to build various models based on these observations. Hence, it is important to remember the bias initially introduced in a study when it concerns data from a test bench.

5.3. Methods summary

Every presented method tries to solve the ageing estimation problem by different manners but each one has its own disadvantages. Current studies focused on models are not able to consider all the existent phenomena to estimate battery ageing. Furthermore, most studies consider only one of the battery ageing phenomena at a time: the capacity fade [146] or the resistance [147] raise. However, it is well known that both have a significant impact on the electric vehicle use (Part 4).

Each of the presented methods can perform well under its own particular conditions: going from a unique detailed battery to a very large and diverse data sets coming from a vehicle fleet. Moreover, chemical studies are not reproducible as are directly dependent on the battery design and its technology. Statistical methods do not need sophisticated measures but large and various data sets in order to find all existing interactions. The main inconvenient here is the complexity to obtain large data due to the time needed to significantly age batteries.

Many studies solve this time problem with accelerated life tests [77,124,148], but this methodology has two main drawbacks. First, an accelerated life test is usually done with a test bench. Hence, the impact of all environmental variables occurring in real life conditions is not taken into account, which produces some errors. Furthermore, these methods cannot perform well enough to obtain the same battery ageing as in real life, due to the lower total storage time but also because of the complex interaction between each variable, which was not considered here.

This diversity of methods induces huge compromises to obtain a generic method performing well for an electric vehicle utilization, especially with the aim of a real time result. Table 1 also illustrates this idea.

The adaptation to electric vehicles requires an easily flexible method, and therefore rejects complicated measurements. Thus, an ideal method would perform quickly with a few and easily obtainable variables. Considering the real time calculus criteria, only two of the presented methods are acceptable: equivalent circuit model and statistical methods. Moreover, the precision level is important for a vehicle use and the best method is direct measurements which are hardly realizable.

5.4. Proposed method

A method that answers to all of these criteria does not currently exist, but it seems that the most promising candidate would be a mix between the measurements precision, the statistical methods adaptability and the understanding of physico-chemical process interactions by modeling. Thus, such method will be accurate for all batteries of the same technology. Furthermore, it will avoid complex measures and permit possible on-line methods for automotive applications. This method could be based on an electrochemical, or a performance model, with complete and non-linear relationships.

Such a method can be updated with data from bench tests and from a vehicle fleet under real usages. This would enable us to obtain many informations from various uses and different data acquisition process which provides a guarantee of the model robustness.

In order to be adaptable on a vehicle context, a method requires to be efficient for all kind of usages. This means a method built from real tests and not from specific bench tests. Furthermore, it is still uncertain if a method without historical update can be performed in a battery diagnosis goal. Hence, very few methods deal with the possibility of a battery diagnosis directly from obtained signals but most consider all the battery history. A proposed idea is to consider the battery performances degradations with an on-line diagnosis approach, without a conservation of its full utilization history. Such method will be only based on the immediate recorded data which is a remarkable advantage.

6. Final conclusion

The lithium-ion battery as the main energy storage solution to the transportation sector is part and parcel of current research. Furthermore, ageing effects occur at each moment of the battery life and are one of the most binding criteria of this technology. In the context of EV, the battery ageing engenders a significant degradation of its performances. These degradations are reflected by an autonomy fade along with a diminution of the acceleration power. These points are particularly constraining for the expansion of the EV market.

This paper presented a review of the battery ageing mechanisms, and their consequences, occurring during a battery life. Different methods tried to explain these phenomena going from an electrochemical point of view to a more data analysis oriented study. Nevertheless, the ageing processes of lithium-ion batteries are complex and strongly dependent on operating conditions. In addition, it is still difficult to quantify the different mechanisms and these mechanisms are correlated and cross-dependent.

These methods coming from a very large set of fields are exposed in this paper. Furthermore, their respective characteristics are discussed and possible new approaches are evoked. Each presented method has advantages and drawbacks, as none of them permits to explore the entire dependencies and correlations of ageing battery factors. There is currently no study considering ageing as a consequence of all the existent interactions between environment and utilization mode. Most of the time, studies only take into consideration the capacity fade in order to define ageing indicators and just a few interpret ageing as a combination of capacity fade and resistance raise. The ageing battery estimation engenders a large set of area, making the problematic really interesting and very challenging.

Therefore, obtaining a complete battery diagnosis based on every ageing factor and compatible with a vehicle use, is still a major remaining challenge. The current focus needs to be set on finding the ideal compromise between developing ageing estimation methods combined with a real time compatibility in order to be accurate. Hence, the final goal of an ageing battery estimation, running in real time for electric vehicles, for all kinds of use, requires various compromises.

References

- [1] T. Nagaura, K. Tozawa, *Progress in Batteries and Solar Cells* 209 (1990) 9.
- [2] R. Brodd, *Comments on the History of Lithium-ion Batteries* (2002).
- [3] T. Kodama, H. Sakaebe, *Journal of Power Sources* 81–82 (1999) 144–149.
- [4] *Technology Roadmap Electric and Plug-in Hybrid Electric Vehicles*, Technical Report, June 2011.
- [5] *Electric Vehicles Roadmap*, Technical Report, 2011.
- [6] J. Neubauer, A. Brooker, E. Wood, *Journal of Power Sources* (2012).
- [7] CPPELEC, *Comparatif de cout d'utilisation d'une renault fluence électrique vs 15 dci 85*, Technical Report, CPPELEC Formation Conseil en Electricité et Energie, 2011.
- [8] R. Faria, P. Moura, J. Delgado, A.T. de Almeida, *Energy Conversion and Management* 61 (2012) 19–30.
- [9] T.J. Miller, *Electrical Energy Storage for Vehicles: TARGETS and Metrics*, Technical Report, U.S. Department of Energy's Advanced Research Projects Agency – Energy (November 2009).
- [10] *Technical Report Electric Vehicle Technology Roadmap for Canada* (2010).
- [11] B. Scrosati, J. Garche, *Journal of Power Sources* 195 (9) (2010) 2419–2430.
- [12] *Plug-in Hybrid Electric Vehicle R&D Plan*, Technical Report, U.S. Department of Energy, February 2007.
- [13] V. Pop, H.J. Bergveld, P.H.L. Notten, P.P.L. Regtien, *Institute of Physics Publishing* 16 (12) (2005) R93–R110.
- [14] J. Vetter, P. Novák, M. Wagner, C. Veit, K.-C. Möller, J. Besenhard, M. Winter, M. Wohlfahrt-Mehrens, C. Vogler, A. Hammouche, *Journal of Power Sources* 147 (1–2) (2005) 269–281.
- [15] F. Joho, P. Novak, M.E. Spahr, *Journal of the Electrochemical Society* 149 (2002) A1020–A1024.
- [16] H. Jannesari, M. Emami, C. Ziegler, *Journal of Power Sources* 196 (22) (2011) 9654–9664.
- [17] Y. Wu, E. Rahm, R. Holze, *Journal of Power Sources* 114 (2) (2003) 228–236.
- [18] P. Arora, R.E. White, M. Doyle, *Journal of the Electrochemical Society* 145 (10) (1998) 3647–3667.
- [19] H. Buqa, A. Würsig, J. Vetter, M. Spahr, F. Krumeich, P. Novák, *Journal of Power Sources* 153 (2) (2006) 385–390. Selected Papers Presented at the 2004 Meeting of the International Battery Association 2004 International Meeting of the International Battery Association.
- [20] P. Balakrishnan, R. Ramesh, T.P. Kumar, *Journal of Power Sources* 155 (2) (2006) 401–414.
- [21] Q. Wang, P. Ping, X. Zhao, G. Chu, J. Sun, C. Chen, *Journal of Power Sources* 208 (2012) 210–224.
- [22] S. Zhang, M.S. Ding, K. Xu, J. Allen, T.R. Jow, *Electrochemical and Solid State Letters* 4 (2001) 206–208.
- [23] D. Goers, M.E. Spahr, A. Leone, W. Märkle, P. Novák, *Electrochimica Acta* 56 (11) (2011) 3799–3808.
- [24] M. Broussely, S. Herreyre, P. Biensan, P. Kasztejna, K. Nechev, R. Staniewicz, *Journal of Power Sources* 97–98 (2001) 13–21. Proceedings of the 10th International Meeting on Lithium Batteries.
- [25] T. Abe, H. Fukuda, Y. Iriyama, Z. Ogumi, *Journal of the Electrochemical Society* 151 (2004) A1120–A1123.
- [26] G.-C. Chung, H.-J. Kim, S.-I. Yu, S.-H. Jun, J. Wook Choi, M.-H. Kim, *Journal of the Electrochemical Society* 147 (2000) 4391–4398.
- [27] S.S. Zhang, *Journal of Power Sources* 162 (2) (2006) 1379–1394. Special issue including selected papers from the International Power Sources Symposium 2005 together with regular papers.
- [28] S. Bourlot, P. Blanchard, S. Robert, *Journal of Power Sources* 196 (16) (2011) 6841–6846, 15th International Meeting on Lithium Batteries (IMLB).
- [29] S. Bashash, S.J. Moura, J.C. Forman, H.K. Fathy, *Journal of Power Sources* 196 (1) (2011) 541–549.
- [30] R.N. Methekar, P.W.C. Northrop, K. Chen, R.D. Braatz, V.R. Subramaniana, *Journal of the Electrochemical Society* 158 (4) (2011) A363–A370.
- [31] M. Koltypin, D. Aurbach, L. Nazar, B. Ellis, *Journal of Power Sources* 174 (2) (2007) 1241–1250, 13th International Meeting on Lithium Batteries.
- [32] S. Zhang, K. Xu, T. Jow, *Journal of Solid State Electrochemistry* 7 (2002) 147–151.
- [33] S. Zhang, K. Xu, T. Jow, *Journal of Power Sources* 115 (1) (2003) 137–140.
- [34] M. Dubarry, C. Truchot, B.Y. Liaw, *Journal of Power Sources* 219 (2012) 204–216.
- [35] P. Liu, J. Wang, J. Hicks-Garner, E. Sherman, S. Soukiazian, M. Verbrugge, H. Tataria, J. Musser, P. Finamore, *Journal of the Electrochemical Society* 157 (4) (2010) A499–A507.
- [36] M. Kassem, J. Bernard, R. Revel, S. Pélissier, F. Duclaud, C. Delacourt, *Journal of Power Sources* 208 (2012) 296–305.
- [37] B.L. Ellis, K.T. Lee, L.F. Nazar, *Chemistry of Materials* 22 (2009) 691–714.
- [38] K. Edstrom, T. Gustafsson, J.O. Thomas, *Electrochimica Acta* 50 (2–3) (2004) 397–403.
- [39] M. Kerlau, M. Marcinek, V. Srinivasan, R.M. Kostecki, *Electrochimica Acta* 52 (17) (2007) 5422–5429.
- [40] K. Xu, A. von Cresce, *Journal of Materials Chemistry* 21 (2011).
- [41] K. Amine, J. Liu, I. Belharouk, *Electrochemistry Communications* 7 (7) (2005) 669–673.
- [42] J. Zhu, K. Zeng, L. Lu, *Electrochimica Acta* 68 (2012) 52–59.
- [43] M. Wohlfahrt-Mehrens, C. Vogler, J. Garche, *Journal of Power Sources* 127 (1–2) (2004) 58–64. Eighth Ulmer Electrochemische Tage.
- [44] D. Abraham, E. Reynolds, E. Sammann, A. Jansen, D. Dees, *Electrochimica Acta* 51 (3) (2005) 502–510.
- [45] Q. Zhang, R.E. White, *Journal of Power Sources* 179 (2) (2008) 793–798.
- [46] J. Li, E. Murphy, J. Winnick, P. Kohl, *Journal of Power Sources* 102 (1–2) (2001) 294–301.
- [47] E. Meissner, G. Richter, *Journal of Power Sources* 144 (2) (2005) 438–460. Selected Papers from the Ninth European Lead Battery Conference Ninth European Lead Battery Conference.
- [48] O. Erdinc, B. Vural, M. Uzunoglu, *A Dynamic Lithium-ion Battery Model Considering the Effects of Temperature and Capacity Fading*, 2009.

- [49] G. Sarre, P. Blanchard, M. Broussely, *Journal of Power Sources* 127 (1–2) (2004) 65–71. Eighth Ulmer Electrochemische Tage.
- [50] A. Ritchie, *Journal of Power Sources* 136 (2) (2004) 285–289. Selected Papers Presented at the International Power Sources Symposium.
- [51] I. Bloom, B. Cole, J. Sohn, S. Jones, E. Polzin, V. Battaglia, G. Henriksen, C. Motloch, R. Richardson, T. Unkelhaeuser, D. Ingersoll, H. Case, *Journal of Power Sources* 101 (2) (2001) 238–247.
- [52] R. Wright, C. Motloch, J. Belt, J. Christophersen, C. Ho, R. Richardson, I. Bloom, S. Jones, V. Battaglia, G. Henriksen, T. Unkelhaeuser, D. Ingersoll, H. Case, S. Rogers, R. Sutula, *Journal of Power Sources* 110 (2) (2002) 445–470.
- [53] R.P. Ramasamy, R.E. White, B.N. Popov, *Journal of Power Sources* 141 (2) (2005) 298–306.
- [54] W. Bögel, J.P. Büchel, H. Katz, *Journal of Power Sources* 72 (1) (1998) 37–42.
- [55] K. Amine, C. Chen, J. Liu, M. Hammond, A. Jansen, D. Dees, I. Bloom, D. Vissers, G. Henriksen, *Journal of Power Sources* 97–98 (2001) 684–687. Proceedings of the 10th International Meeting on Lithium Batteries.
- [56] S. Zhang, K. Xu, T. Jow, *Electrochimica Acta* 49 (7) (2004) 1057–1061.
- [57] K. Ohue, T. Utsunomiya, O. Hatozaki, N. Yoshimoto, M. Egashira, M. Morita, *Journal of Power Sources* 196 (7) (2011) 3604–3610. cited by (since 1996) 2.
- [58] M. Broussely, P. Biensan, F. Bonhomme, P. Blanchard, S. Herreyre, K. Nechev, R. Staniewicz, *Journal of Power Sources* 146 (1–2) (2005) 90–96. Selected Papers Presented at the 12th International Meeting on Lithium Batteries, 12th International Meeting on Lithium Batteries.
- [59] K. Nunotani, F. Yoshida, Y. Kamiya, Y. Daisho, K. Abe, M. Kono, H. Matsuo, Development and Performance Evaluation of Lithium Iron Phosphate Battery with Superior Rapid Charging Performance, in: *Vehicle Power and Propulsion Conference (VPPC)*, IEEE, 2011, pp. 1–4.
- [60] J. Gnanaraj, E. Zinigrad, L. Asraf, H. Gottlieb, M. Sprecher, D. Aurbach, M. Schmidt, *Journal of Power Sources* 119–121 (2003) 794–798. Selected Papers Presented at the 11th International Meeting on Lithium Batteries.
- [61] H. Yang, H. Bang, K. Amine, J. Prakash, *Journal of the Electrochemical Society* 152 (1) (2005) A73–A79.
- [62] S.B. Peterson, J. Apt, J. Whitacre, *Journal of Power Sources* 195 (8) (2010) 2385–2392.
- [63] J.R. Belt, C.D. Ho, C.G. Motloch, T.J. Miller, T.Q. Duong, *Journal of Power Sources* 123 (2) (2003) 241–246.
- [64] R. Kötz, P. Ruch, D. Cericola, *Journal of Power Sources* 195 (3) (2010) 923–928.
- [65] K. Asakura, M. Shimomura, T. Shodai, *Journal of Power Sources* 119–121 (2003) 902–905. Selected Papers Presented at the 11th International Meeting on Lithium Batteries.
- [66] H. Gong, Y. Yu, T. Li, T. Mei, Z. Xing, Y. Zhu, Y. Qian, X. Shen, *Materials Letters* 66 (1) (2012) 374–376.
- [67] M. Dubarry, B.Y. Liaw, *Journal of Power Sources* 194 (1) (2009) 541–549. cited by (since 1996) 16.
- [68] Y. Zhang, C.-Y. Wang, X. Tang, *Journal of Power Sources* 196 (3) (2011) 1513–1520.
- [69] D. Doerffel, S.A. Sharkh, *Journal of Power Sources* 155 (2) (2006) 395–400.
- [70] M. Conte, F.V. Conte, I. Bloom, K. Morita, T. Ikeya, J. Belt, Ageing Testing Procedures on Lithium Batteries in an International Collaboration Context, 2010.
- [71] A. Widodo, M.-C. Shim, W. Caesarendra, B.-S. Yang, *Expert Systems with Applications* 38 (9) (2011) 11763–11769.
- [72] Y. Xing, K.-L. Tsui, N. Williard, M. Pecht, A Comparative Review of Prognostics-Based Reliability Methods for Lithium Batteries, in: *Prognostics and System Health Management Conference (PHM-Shenzhen)*, 2011, pp. 1–6.
- [73] E. Meissner, G. Richter, *Journal of Power Sources* 116 (1–2) (2003) 79–98. Selected Papers Presented at the Eighth European Lead Battery Conference.
- [74] C. Hu, B.D. Youn, J. Chung, *Applied Energy* (2011).
- [75] Y.-H. Chiang, W.-Y. Sean, J.-C. Ke, *Journal of Power Sources* 196 (8) (2011) 3921–3932.
- [76] Q. Zhang, R.E. White, *Journal of Power Sources* 179 (2) (2008) 785–792.
- [77] M. Ecker, J.B. Gerschler, F. Vogel, S. Käbitz, F. Hust, P. Dechent, D.U. Sauer, *Journal of Power Sources* (2012).
- [78] M. Doyle, T.F. Fuller, J. Newman, *Journal of the Electrochemical Society* 140 (6) (1993) 1526–1533.
- [79] T.F. Fuller, M. Doyle, J. Newman, *Journal of the Electrochemical Society* 141 (1) (1994) 1–10.
- [80] M. Doyle, J. Newman, *Journal of Power Sources* 54 (1) (1995) 46–51.
- [81] M. Doyle, J. Newman, A.S. Gozdz, C.N. Schmutz, J.-M. Tarascon, *Journal of the Electrochemical Society* 143 (6) (1996) 1890–1903.
- [82] J. Newman, W. Tiedemann, *AIChE Journal* 21 (1) (1975) 25–41.
- [83] R. Darling, J. Newman, *Journal of the Electrochemical Society* 145 (3) (1998) 990–998.
- [84] J. Christensen, J. Newman, *Journal of the Electrochemical Society* 150 (11) (2003) A1416–A1420.
- [85] J. Christensen, J. Newman, *Journal of the Electrochemical Society* 151 (11) (2004) A1977–A1988.
- [86] J. Christensen, J. Newman, *Journal of the Electrochemical Society* 153 (6) (2006) A1019–A1030.
- [87] J. Christensen, J. Newman, *Journal of Solid State Electrochemistry* 10 (5) (2006) 293–319.
- [88] H.J. Ploehn, P. Ramadass, R.E. White, *Journal of the Electrochemical Society* 151 (3) (2004) A456–A462.
- [89] V. Ramadesigan, V. Boovaragavan, J.C. Pirkle, V.R. Subramanian, *Journal of the Electrochemical Society* 157 (7) (2010) A854–A860.
- [90] V. Ramadesigan, K. Chen, N.A. Burns, V. Boovaragavan, R.D. Braatz, V.R. Subramanian, *Journal of the Electrochemical Society* 158 (9) (2011) A1048–A1054.
- [91] M. Safari, C. Delacourt, *Journal of the Electrochemical Society* 158 (2) (2011) A62–A73.
- [92] M. Safari, C. Delacourt, Simulation-based Analysis of Aging Phenomena in a Commercial Graphite/LiFePO₄ Cell vol. 158, *Electrochemical Society*, Pennington, NJ, ETATS-UNIS, 2011.
- [93] M. Safari, C. Delacourt, *Journal of the Electrochemical Society* 158 (10) (2011) A1123–A1135.
- [94] A.L. Dalverny, J.S. Filhol, M.L. Doublet, *Journal of Materials Chemistry* 21 (27) (2011) 10134–10142.
- [95] M. Wagemaker, D.P. Singh, W.J.H. Borghols, U. Lafont, L. Haverkate, V.K. Peterson, F.M. Mulder, *Journal of the American Chemical Society* 133 (26) (2011) 10222–10228.
- [96] K. Tasaki, A. Goldberg, J.-J. Lian, M. Walker, A. Timmons, S.J. Harris, *Journal of the Electrochemical Society* 156 (12) (2009) A1019–A1027.
- [97] K. Tasaki, S.J. Harris, *The Journal of Physical Chemistry C* 114 (17) (2010) 8076–8083.
- [98] K. Leung, J.L. Budzien, *Physical Chemistry Chemical Physics* 12 (25) (2010) 6583–6586.
- [99] D. Haifeng, W. Xuezhong, S. Zechang, A New SOH Prediction Concept for the Power Lithium-ion Battery Used on HEVs, in: *Vehicle Power and Propulsion Conference*, 2009, VPPC '09, IEEE, 2009, pp. 1649–1653.
- [100] M. Einhorn, V. Conte, C. Kral, J. Fleig, R. Permann, Parameterization of an Electrical Battery Model for Dynamic System Simulation in Electric Vehicles, in: *Vehicle Power and Propulsion Conference (VPPC)*, 2010 IEEE, 2010, pp. 1–7.
- [101] A. Eddahech, O. Briat, H. Henry, J.-Y. Delétage, E. Woïrgard, J.-M. Vinassa, *Microelectronics Reliability* 51 (9–11) (2011) 1968–1971. Proceedings of the 22th European Symposium on the Reliability of Electron Devices, Failure Physics and Analysis.
- [102] A. Eddahech, O. Briat, E. Woïrgard, J. Vinassa, *Microelectronics Reliability* 52 (9–10) (2012) 2438–2442. Special Issue 23rd European Symposium on the Reliability of Electron Devices, Failure Physics and Analysis.
- [103] D. Andre, C. Appel, T. Soczka-Guth, D.U. Sauer, *Journal of Power Sources* 224 (2013) 20–27.
- [104] V. Vapnik, Springer-Verlag, New York, 1995.
- [105] M.E. Tipping, *Journal of Machine Learning Research* 1 (2001) 211–244.
- [106] C.M. Bishop, *Pattern Recognition and Machine Learning*, first ed., Springer, 2006.
- [107] B. Saha, K. Goebel, S. Poll, J. Christophersen, *IEEE Transactions on Instrumentation and Measurement* 58 (2) (2009) 291–297.
- [108] B. Saha, K. Goebel, S. Poll, J. Christophersen, An Integrated Approach to Battery Health Monitoring Using Bayesian Regression and State Estimation, in: *Autotestcon*, 2007 IEEE, 2007, pp. 646–653.
- [109] K. Smith, G.-H. Kim, A. Pesaran, Modeling of Nonuniform Degradation in Large-format Li-ion Batteries. Presented at the 215th Electrochemical Society Meeting, vol. 25–29 May 2009, San Francisco, CA.
- [110] U. Tröltzsch, O. Kanoun, H.-R. Tränkler, *Electrochimica Acta* 51 (8–9) (2006) 1664–1672. Electrochemical Impedance Spectroscopy Selection of papers from the 6th International Symposium (EIS 2004) 16–21 May 2004, Cocoa Beach, FL, USA.
- [111] B.Y. Liaw, R.G. Jungst, G. Nagasubramanian, H.L. Case, D.H. Doughty, *Journal of Power Sources* 140(1) (2005) 157–161.
- [112] J. Belt, V. Utgikar, I. Bloom, *Journal of Power Sources* 196 (23) (2011) 10213–10221.
- [113] S.S. Choi, H.S. Lim, *Journal of Power Sources* 111 (1) (2002) 130–136.
- [114] B.Y. Liaw, E. Roth, R.G. Jungst, G. Nagasubramanian, H.L. Case, D.H. Doughty, *Journal of Power Sources* 119–121 (2003) 874–886. Selected Papers Presented at the 11th International Meeting on Lithium Batteries.
- [115] E. Thomas, I. Bloom, J. Christophersen, V. Battaglia, *Journal of Power Sources* 184 (1) (2008) 312–317.
- [116] H. Wenzl, I. Baring-Gould, R. Kaiser, B.Y. Liaw, P. Lundsager, J. Manwell, A. Ruddell, V. Svoboda, *Journal of Power Sources* 144 (2) (2005) 373–384. Selected Papers from the Ninth European Lead Battery Conference.
- [117] P. Ramadass, B. Haran, R. White, B.N. Popov, *Journal of Power Sources* 123 (2) (2003) 230–240.
- [118] J. Wang, P. Liu, J. Hicks-Garner, E. Sherman, S. Soukiazian, M. Verbrugge, H. Tataria, J. Musser, P. Finamore, *Journal of Power Sources* 196 (8) (2011) 3942–3948.
- [119] O. Bohlen, J. Kowal, D.U. Sauer, *Journal of Power Sources* 173 (1) (2007) 626–632.
- [120] S. Sankarasubramanian, B. Krishnamurthy, *Electrochimica Acta* 70 (2012) 248–254.
- [121] S. Mishra, M. Pecht, T. Smith, I. McNeer, R. Harris, Remaining Life Prediction of electronic Products Using Life Consumption Monitoring Approach, in: *European Microelectronics Packaging and Interconnection Symposium*, 2002.
- [122] V. Marano, S. Onori, Y. Guezennec, G. Rizzoni, N. Madella, Lithium-ion Batteries Life Estimation for Plug-in Hybrid Electric Vehicles, in: *Vehicle Power and Propulsion Conference*, 2009, VPPC '09, IEEE, 2009, pp. 536–543.
- [123] M. Safari, M. Morcrette, A. Teyssot, C. Delacourt, *Journal of the Electrochemical Society* 157 (6) (2010) A713–A720.

- [124] K. Takei, K. Kumai, Y. Kobayashi, H. Miyashiro, N. Terada, T. Iwahori, T. Tanaka, *Journal of Power Sources* 97–98 (2001) 697–701. Proceedings of the 10th International Meeting on Lithium Batteries.
- [125] C. Chen, M. Pecht, *Prognostics of Lithium-ion Batteries Using Model Based and Data-driven Methods*, 2012.
- [126] K.S. Ng, C.-S. Moo, Y.-P. Chen, Y.-C. Hsieh, *Applied Energy* 86 (9) (2009) 1506–1511.
- [127] T. Hansen, C.-J. Wang, *Journal of Power Sources* 141 (2) (2005) 351–358.
- [128] L.A. Zadeh, *Information Sciences* 178 (13) (2008) 2751–2779.
- [129] A. Zenati, P. Desprez, H. Razik, Estimation of the SOC and the SOH of Li-ion Batteries, by Combining Impedance Measurements with the Fuzzy Logic Inference, in: *IECON 2010–36th Annual Conference on IEEE Industrial Electronics Society*, 2010, pp. 1773–1778.
- [130] K. Tsang, W. Chan, *Energy Conversion and Management* 65 (2013) 7–12.
- [131] A.J. Salkind, C. Fennie, P. Singh, T. Atwater, D.E. Reisner, *Journal of Power Sources* 80 (1–2) (1999) 293–300.
- [132] P. Singh, R. Vinjamuri, X. Wang, D. Reisner, *Electrochimica Acta* 51 (8–9) (2006) 1673–1679. Selection of Papers from the 6th International Symposium (EIS 2004) 16–21 May 2004, Cocoa Beach, FL, USA.
- [133] I.-S. Kim, *IEEE Transactions on Power Electronics* 23 (4) (2008) 2027–2034.
- [134] G.L. Plett, *Journal of Power Sources* 134 (2) (2004) 277–292.
- [135] M. Urbain, *Modelisation électrique et énergétique des accumulateurs lithium ion estimation en ligne du SOC et du SOH*. Ph.D. thesis, INP Lorraine, 2009.
- [136] M. Charkhgard, M. Farrokhi, *IEEE Transactions on Industrial Electronics* 57 (12) (2010) 4178–4187.
- [137] A. Eddahech, O. Briat, N. Bertrand, J.-Y. Delétage, J.-M. Vinassa, *International Journal of Electrical Power & Energy Systems* 42 (1) (2012) 487–494.
- [138] D. Andre, A. Nuhic, T. Soczka-Guth, D. Sauer, *Engineering Applications of Artificial Intelligence* (2012).
- [139] K.-H. Chao, J.-W. Chen, *Expert Systems with Applications* 38 (12) (2011) 15183–15193.
- [140] W. Shen, *Energy Conversion and Management* 48 (2) (2007) 433–442.
- [141] J. Lampinen, A. Vehtari, *Neural Networks* 14 (3) (2001) 257–274.
- [142] G.M. Jenkins, G.E.P. Box, *Time Series Analysis: Forecasting and Control*, Prentice Hall, 1994.
- [143] J. Kozłowski, *Electrochemical Cell Prognostics Using Online Impedance Measurements and Model-based Data Fusion Techniques*, in: *IEEE Aerospace Conference* vol. 7, 2003, pp. 3257–3270.
- [144] W. Weibull, *Journal of Applied Mechanics* (1951) 293–297.
- [145] S.-W. Eom, M.-K. Kim, I.-J. Kim, S.-I. Moon, Y.-K. Sun, H.-S. Kim, *Journal of Power Sources* 174 (2) (2007) 954–958, 13th International Meeting on Lithium Batteries.
- [146] Ramadass Haran, Popov White, Capacity Fade of Li-ion Cells Cycled at Different Temperatures, in: *Battery Conference on Applications and Advances*, 2002, pp. 13–18.
- [147] H. Schranzhofer, J. Bugajski, H.J. Santner, C. Korepp, K.C. Moller, J.O. Besenhard, M. Winter, W. Sitte, *Journal of Power Sources* 153 (2) (2006) 391–395.
- [148] O. Bohlen, J. Kowal, D.U. Sauer, *Journal of Power Sources* 172 (1) (2007) 468–475. ACS San Francisco 2006 Fuel and Cell Symposium, American Chemical Society National Meeting, San Francisco, CA Sept 10–14 2006.