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Review

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

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HIGHLIGHTS

- 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.

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ABSTRACT

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.

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

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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

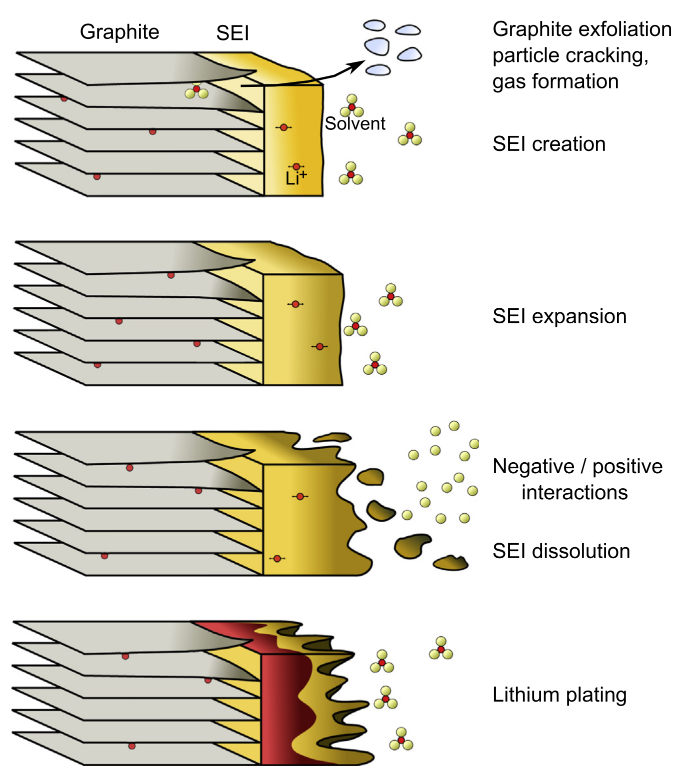

Fig. 1. Illustration of ageing effects on battery negative electrode: the capacity fade and the SEI raise.