Journal of Power Sources 325 (2016) 273–285

Contents lists available at ScienceDirect

Journal of Power Sources

journal homepage: www.elsevier.com/locate/jpowsour

Lithium battery aging model based on Dakin's degradation approach

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HIGHLIGHTS

- Study of calendar and power cycling aging under different aging conditions for two different battery technologies.
- The logarithms of resistance rise and battery capacity fade evolve linearly with time.
- Battery capacity degradation rate fulfill Eyring's law which is extended to take into account current magnitude effect.
- Battery aging rate depend on current, temperature, battery state of charge and is defined in one expression.
- Two different aging behaviors noticed for the ranges of −5 °C to 25 °C and 25 °C to 60 °C.

ARTICLE INFO

Article history:
Received 23 February 2016
Received in revised form
3 May 2016
Accepted 8 June 2016

Keywords:
Lithium battery
Calendar aging
Power cycling
Ageing model
Eyring's law
Dakin's law

ABSTRACT

This paper proposes and validates a calendar and power cycling aging model for two different lithium battery technologies. The model development is based on previous SIMCAL and SIMSTOCK project data. In these previous projects, the effect of the battery state of charge, temperature and current magnitude on aging was studied on a large panel of different battery chemistries. In this work, data are analyzed using Dakin's degradation approach. In fact, the logarithms of battery capacity fade and the increase in resistance evolves linearly over aging. The slopes identified from straight lines correspond to battery aging rates. Thus, a battery aging rate expression function of aging factors was deduced and found to be governed by Eyring's law. The proposed model simulates the capacity fade and resistance increase as functions of the influencing aging factors. Its expansion using Taylor series was consistent with semi-empirical models based on the square root of time, which are widely studied in the literature. Finally, the influence of the current magnitude and temperature on aging was simulated. Interestingly, the aging rate highly increases with decreasing and increasing temperature for the ranges of −5 °C–25 °C and 25 °C–60 °C, respectively.

1. Introduction

Lithium batteries are key solutions as power storage systems for several applications including portable devices, aviation, space, and electrified vehicles. Their success is principally due to their high power and energy density [1]. Therefore, several researchers are attempting to develop more powerful, cheaper, longer-lived and more secure batteries [1–5].

One drawback of lithium batteries is their durability: lithium batteries' energy and power capability decrease over time. The degradation rate is sensitive to operating conditions. In fact, battery temperature, state of charge (SOC), voltage, depth of discharge (DOD), and current magnitude are the most influential and studied aging factors [6–8,47].

The main aging processes are related to, but not limited to, solid electrolyte interphase (SEI) growth, active material loss, and lithium plating [6,9–11].

A crucial step towards the large-scale introduction of electrified vehicles in the market is to reduce the cost of their energy storage devices. In fact, a battery pack represents nearly half the price of an electric vehicle. Some vehicle manufacturers rent battery packs to the end users and guarantee their replacement if they no longer meet the minimal required performance. The vehicle manufacturer assumes an economical risk and must be sure of the profitability of

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http://dx.doi.org/10.1016/j.jpowsour.2016.06.036
0378-7753/

this operation. Thus, modeling the evolution of battery pack performance over time facilitates the discovery of strategies to enhance its durability and properly manage the warranty.

Aging models should be compatible with vehicle usage. Generally, vehicles spend most of their time (90%) unused or parked at a random and variable aging condition. Thus, calendar aging rate should take into account temperature and battery SOC (or voltage) as thermo-oxidative aging factors. Likewise, power cycling highly contributes to the total aging. In real applications, the power profile consists of successive charges and discharges that are completely random and depend on individual's driving style. At a laboratory scale, testing a battery as in actual use is a difficult task. Thus, relevant accelerated aging tests are crucial to calibrate the models. An optimal design of experiment could save time and resources but cannot cover all possible and imaginable operating conditions.

Several models have been developed for (but not limited to) this purpose and are based on different approaches [8,12–14]. Lithium battery aging models could be classified to semi-empirical [7,8,15,48], fatigue approach [13,16,17], and electrochemical models [14,18,19]. This is only a short literature overview of available aging models, which are discussed and compared according to their classification.

Semi-empirical models are widely studied in the literature, and their structure highly depends on the design of experiments. Some specific examples are discussed below.

In the SIMCAL project, the calendar aging of different batteries was principally studied under 3 different temperatures (30, 45, 60 °C) and SOCs (30, 65, 100%) for a total of 9 different aging conditions [7,20]. After each performance checkup, the aging battery SOC was set by discharging the same number of ampere-hours after a full battery charge. The batteries were then set in climatic chambers at the aging temperature without voltage control. Consequently, the battery self-discharge could not be mastered during aging. Likewise, a drift is noted in the SOC of aging batteries as the capacity decreases during aging [21].

Schmalstieg et al. studied calendar aging at 12 different SOCs (0–100%) only under 50 °C and at 50% of SOC for 3 different temperatures (35, 40, 50 °C) to establish the Arrhenius law parameters (activation energy, pre-exponential constant) [15]. Thus, temperature effect on aging was deduced using the same parameters for the remaining SOCs. In contrast to the SIMCAL approach, the battery voltage was maintained during aging. In fact, the battery SOCs were set from the OCV relation to the SOC, and the calendar model considers the voltage as an aging factor instead of the SOC. This different approach allows the aging conditions to be mastered and to avoid ambiguities. However, maintaining the battery voltage disturbs the thermodynamic equilibrium of the battery as it exchanges continuously with the voltage source.

Wang et al. studied lithium battery aging at several DODs (10–90%), temperatures (10–43 °C) and current rates (C/2 to 6.5 C) for a total of 60 different aging conditions [8]. The tests under low current (C/2) and DOD (10%) were assumed to represent the calendar aging. This is a strong assumption because C/2 current is not too low and corresponds to the ranges of currents used in electric vehicle applications. The principal disadvantage of this approach is that it does not consider battery SOC as an aging factor, which highly influences the battery aging rate.

These different approaches lead to a different calibration of the calendar aging models. Some assumptions are required to optimize the number of tests or to master aging conditions. Indeed, this could lead to different aging behaviors. Nevertheless, the battery capacity fade and resistance increase were found to evolve according to the square root of time for the different discussed approaches.

The power cycling aging model developed by Wang et al. considers temperature, DOD, and current magnitude as aging factors [8]. To build the power cycling aging model, the contribution of calendar aging to the total aging was subtracted. Thus, the capacity losses were proportional to ampere-hour (Ah) throughput, and the aging rate increases with increasing DOD, current magnitude and temperature.

Schmalstieg et al. tested the aging of batteries under power cycling at only one current magnitude (1 C) and one temperature (35 °C) [15]. In fact, the batteries were cycled among different average SOCs (average voltages) with different ΔSOC (22 different aging conditions). To build the power cycling model, the contribution of calendar aging to the total aging was subtracted as executed by Wang et al. [8]. The pure power cycling model considers only the average voltage and ΔSOC as aging factors. The temperature contribution to the total aging was represented only in the calendar part of the total aging model. On the other hand, capacity losses due to power cycling were proportional to the square root of Ah-throughput, and the aging rate increases with increasing ΔSOC, average voltage, but not sensitive to temperature.

Ah throughput representation could lead to ambiguities regarding how to separate calendar and power cycling contribution. In fact, the total aging is described with a double representation, over time and Ah-throughput for calendar and power cycling aging, respectively. In our point of view, the challenge is how to represent the total aging function of time with only one aging rate expression that ensures the continuity between the two aging modes. This could answer many ambiguities and may facilitate the degradation rate identification.

The fatigue approach models are generally based on the Palmgren–Miner theory used for mechanical components [22,23] and could be coupled to a special damage counting technique called rainflow counting [24] and/or to machine learning methods as support vector machines or neural networks [13,49]. The rainflow counting technique is useful to discretize a random electric power solicitation to micro cycles that could be studied independently at a laboratory scale. The principal advantage of fatigue models is their compatibility with the cumulative degradation approach.

These models are generally based on the relation between the number of cycles and the failure function of DOD. Some corresponding models consider only the DOD as an aging factor [25,26]. Thus, the swapping SOCs are counted during power cycling, classified in different DOD magnitudes with corresponding time durations. The battery degradation is then accumulated, and battery performance evolution over time is deduced. Some other works combine the effects of DOD, temperature and current magnitude [27,28]. The principal drawback of these models is their inconsistency in representing calendar aging, especially when all aging factors are constant.

Nuhic et al. coupled the rainflow counting technique with support vector machines [13]. Under this learning method, the model parameters are updated and optimized over time, allowing prognostics for other power cycling profiles. Otherwise, Safari et al. coupled the fatigue approach to porous electrode and chemical kinetics theory. Dudézert et al. developed their aging model by an analogy between mechanical load and electrochemical load [16].

In contrast, the electrochemical approach describes battery aging from the perspective of chemical kinetics. Safari et al. proposed a multimodal physics-based model to describe battery aging [14]. In fact, the battery was modeled using porous electrode model theory [29]. Thus, battery aging was described through SEI film growth, which is assumed to be proportional to side-reaction current and determined from material balance of solvents. Safari et al.'s approach is very interesting as an electrochemical interpretation of aging phenomena. In fact, it could be used to optimize battery