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A Comparison between Electrochemical Impedance Spectroscopy and Incremental Capacity-Differential Voltage as Li-ion Diagnostic Techniques to Identify and Quantify the Effects of Degradation Modes within Battery Management Systems

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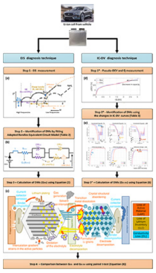
HIGHLIGHTS

- Degradation modes (DMs) evaluated within parallel connected cells.
- A novel method to quantify the effect of DMs using EIS and IC-DV is presented.
- LLI, LAM are the most pertinent DMs obtained with each technique.
- The effect of the DMs obtained with EIS and IC-DV are correlated.
- On-board implementation of EIS and IC-DV within a BMS is discussed.

GRAPHICAL ABSTRACT



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ABSTRACT

Degradation of Lithium-ion batteries is a complex process that is caused by a variety of mechanisms. For simplicity, ageing mechanisms are often grouped into three degradation modes (DMs): conductivity loss (CL), loss of active material (LAM) and loss of lithium inventory (LLI). State of Health (SoH) is typically the parameter used by the Battery Management System (BMS) to quantify battery degradation based on the decrease in capacity and the increase in resistance. However, the definition of SoH within a BMS does not currently include an indication of the underlying DMs causing the degradation. Previous studies have analysed the effects of the DMs using incremental capacity and differential voltage (IC-DV) and electrochemical impedance spectroscopy (EIS). The aim of this study is to compare IC-DV and EIS on the same data set to evaluate if both techniques provide similar insights into the causes of battery degradation. For an experimental case of parallelized cells aged differently, the effects due to LAM and LLI were found to be the most pertinent, outlining that both techniques are correlated. This approach can be further implemented within a BMS to quantify the causes of battery ageing which would support battery lifetime control strategies and future battery designs.

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1. Introduction

In the recent years, battery electric and plug hybrid electric vehicles (BEV and PHEV) have been presented as an alternative

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road transport mode to conventional internal combustion engine (ICE) based vehicles due to their high energy efficiency and low tail pipe emissions. Vehicle battery systems are typically equipped with a high number of cells connected electrically in parallel and series to meet the requirements of energy and power. During battery life, the available energy and power that may be extracted from the battery is known to reduce due to degradation. Degradation of lithium-ion batteries (LIBs) is an extremely complex process that depends on a variety of ageing mechanisms caused by different intrinsic and extrinsic factors [1,2]. Intrinsic factors include inconsistencies in manufacturing processes and in the materials used. Intrinsic factors are currently mitigated by improving quality control, manufacturing processes and battery designs. Extrinsic factors include those due to the inhomogeneous operating conditions that a LIB may be subject to, e.g. non-uniform current or temperature distribution within the complete battery pack. In order to reduce battery degradation, the Battery Management System (BMS) mitigates the impact of extrinsic factors by setting a number of variables that include, but are not constrained to: the level of charge or discharge power, the temperature range that the battery operates over and the allowable depth of discharge (DoD) of the battery. State of Health (SoH) is typically the parameter used by the BMS to quantify battery degradation with respect to its nominal state and it is often quantified based on two measures: capacity fade (CF) and power fade (PF) [3]. These metrics are directly related to available driving range and power, respectively. However, the definition of SoH within a BMS does not currently include an indication of the underpinning ageing mechanisms causing the degradation.

There are many different ageing mechanisms and to aid in their understanding and interpretation, they are commonly grouped into three different degradation modes (DMs): conductivity loss (CL), loss of active material (LAM) and loss of lithium inventory (LLI) [4]. CL includes the degradation of the electronic parts of the battery such as current collector corrosion or binder decomposition [5]. LAM is related to structural transformations in the active material and electrolyte decomposition [5]. LLI is attributed to the variation of the number of lithium-ions (Li-ions) that are available for intercalation and de-intercalation processes [5].

Several techniques are commonly applied and reported within the literature to identify and quantify the effects of DMs. These are often classified into in-situ and ex-situ electrochemical techniques. In-situ electrochemical methods are not-invasive characterisation techniques, potentially making them suitable for real-time applications within a BMS. Examples of in-situ methods are Incremental Capacity (IC) and Differential Voltage (DV) [2,4,6], Electrochemical Impedance Spectroscopy (EIS) [7,8,9] and Differential Thermal Voltammetry (DTV) [10,11].

Ex-situ methods consist of applying physicochemical and electrochemical invasive techniques to study the cells internally. Scanning Electron Microscopy (SEM), Energy Dispersive Spectrometry (EDS) or X-Ray Diffractometry (XRD) [12] are commonly used examples. It is beyond the scope of this paper to discuss each of these techniques; they are however described fully in a number of references [12].

In-situ health diagnosis techniques were shown to be effective tools to analyse DMs of single Li-ion cells. In automotive applications, however, the majority of battery pack configurations connect cells in parallel first to form small modules, and then align the modules in series to form the pack [13]. The main difference between a single cell and a module that has cells connected in parallel is the existence of uneven current distribution when cell properties change due to manufacturing tolerances or usage conditions [3]. For instance, the presence of temperature gradients or different resistance paths within an automotive battery pack will lead to uneven current distribution in the short-term and to cell-to-cell SoH differences in the long-term [14]. Another application of this study is second life grid energy storage applications in which battery modules (of different SoH) may be connected together either in series or parallel to form the complete battery assembly [3]. Understanding the reasons for battery ageing under real operating conditions is needed to improve lifetime control strategies within BMSs and the design of new batteries and manufacturing processes, so that the impact of intrinsic and extrinsic factors on battery ageing can be better mitigated.

From a review of the published literature the authors have identified that two suitable techniques to identify and quantify the effects of DMs are EIS [8,15] and IC-DV [2,4] because they can infer the effects of the different DMs in a mechanistic way and thus, they can be implemented on-board in future BMS real-time applications [16–19]. In line with this, the contribution of this work is twofold. Firstly, a step-by-step methodology to identify and quantify the effects of the DMs is proposed as illustrates in Fig. 1. EIS and IC-DV techniques based on full cell measurements were used to identify and quantify the DMs. Secondly, the results obtained from each technique are critically evaluated and compared within the context of their on-board implementation within a BMS application. To make the analysis of these techniques close to a real application, this study considers the data set from four cells connected in parallel emulating an imbalanced battery module scenario. Each cell's initial SoH was different, which is a typical scenario when battery ageing may ultimately cause a failure in a module due to uneven current distribution through each cell connected in parallel.

The structure of this work is divided as follows: Section 2 explains the most common DMs and ageing mechanisms in LIBs, focusing on the Nickel Cobalt Aluminum - Carbon (NCA-C) cell type. Section 3 gives a background of the EIS and IC-DV diagnosis techniques, describing the methodology employed to identify and quantify the effects of DMs. Section 4 summarises the experimental investigation conducted for this work and Section 5 presents the results. Using the approach described in Section 3, Section 6 identifies and quantifies the DMs, and their relationship to SoC and initial degradation of each cell connected in parallel. Furthermore, the results obtained with EIS and IC-DV are compared and their applicability for embedded use with a BMS are discussed. Section 7 describes the limitations of this work, outlining areas that need to be further investigated and finally, Section 8 presents the main conclusions of this study.

2. Degradation modes and ageing mechanisms in lithium-ion batteries

NCA-C cells are a candidate solution for BEV and PHEV applications due to their high operating voltage window (2.5–4.2 V), high specific discharge capacity (155 mAh g^{-1}) and high specific energy (200–260 Wh kg^{-1}) [20]. The most relevant drawbacks are reliability problems at high temperatures (>40 °C) and relatively high cost of the bulk cobalt (circa: 0.03 £ g^{-1} - 2016) [21] that forms the cell cathode.

From an electrochemical viewpoint, the general causes that lead to CL, LLI or LAM can be very diverse as illustrated in Fig. 1 (c). LLI ageing mechanisms are electrolyte decomposition, lithium plating and formation of Li-ion grains [1]. CL ageing mechanisms are current collector corrosion and binder decomposition [1]. LAM ageing mechanisms are oxidation of the electrolyte, electrode decomposition, intercalation gradient strains in the active particles, and crystal structure disorder [1].

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