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Abstract—**Electric vehicles (EVs) represent one of the solutions for reducing the carbon emissions worldwide. Even though EVs have recently gained more and more popularity, their adoption at a large scale is mainly prevented by several factors, such as range anxiety and battery degradation. The range of an EV is mainly limited by the energy density and specific energy of the battery, while the battery degradation is determined by the driving manner (i.e., the mission profile) to which the EV is subjected to. In this paper we analyze the EV-battery degradation, in terms of both capacity fade and internal resistance increase (power decrease), by performing laboratory accelerated ageing tests for a period of eleven months. To perform this analysis, we used a standardized driving cycle – the Worldwide harmonized Light vehicles Test Cycle (WLTC) and a real-life temperature profile, characteristic to a European city. Furthermore, the study is performed for a Lithium-ion battery chemistry, which is nowadays very popular for EVs, the nickel manganese cobalt oxide-chemistry.**

Keywords—electric vehicle, Lithium-ion battery, driving cycle, accelerated ageing, capacity, internal resistance

I. INTRODUCTION

Lithium-ion (Li-ion) batteries have developed as the key energy storage technology for the transportation sector – mainly for electrical vehicles (EVs), hybrid EVs, and plug-in hybrid EVs [1]. This has become possible due to the continuous development of this technology, which nowadays is characterized by superior features in terms of specific energy, lifetime, and efficiency in comparison to their competitors (i.e., lead-acid batteries, NiMH batteries). However, in comparison to the traditional internal combustion engine vehicles, EVs powered by Li-ion batteries are inferior in terms of energy density (range) and lifetime. Consequently, potential owners/ users become anxious because of the EVs' range and performance while ageing [2].

The Li-ion battery energy storage technology comprises a wide variety of chemistries, which are available to the OEMs. Nevertheless, among these different chemistries, nickel manganese cobalt oxide (NMC) and nickel cobalt aluminum oxide (NCA) are matching the best the requirements of EV batteries since they are characterized by high energy density and high specific energy, and long lifetime [3]. Consequently,

the work carried out in this paper is performed for an NMC-based battery cell.

The two parameters that are describing the performance of an EV-battery are the battery capacity and internal resistance; the capacity of the battery defines the range of the EV, while the internal resistance defines the acceleration. Both parameters are highly dependent on the temperature and on the load current/ power [4]. Furthermore, the capacity and the resistance are degrading in time while the battery is ageing [5] – [7]. Consequently, the range and the acceleration of the EV will worsen during long term use, bringing anxiety to the EV users and prospective users.

Therefore, in this paper we investigated how the capacity and the internal resistance of an NMC-based Li-ion battery cell are degrading in time by performing laboratory ageing tests. In order to obtain realistic degradation behavior of the two parameters, we have applied to the batteries a load current corresponding to a standardized EV driving cycle profile. Moreover, the batteries were aged using the annual temperature profile for the city of Seville, Spain. The aging process lasted for eleven months and after each month of ageing it was performed a check-up for verifying the performance of the NMC-based Li-ion battery cells.

II. STATE-OF-THE-ART

Different aspects are presented in the literature regarding the ageing and lifetime of Li-ion batteries used in EV applications.

The capacity and power degradation behavior of a Li-ion battery (LFP chemistry) used in an EV are studied in [8]; the authors used a semi-empirical lifetime model to determine the expected lifetime of the battery in the case when the EV follows the New European Driving Cycle (NEDC) profile [8]. Nonetheless, the authors considered a constant temperature during the simulations and no laboratory experiments were carried out using the NEDC profile.

In [9], Marano et al., have estimated the lifetime of a generic Li-ion battery used in plug-in hybrid electric vehicle (PHEV); the authors have used a simple lifetime model (based on charge throughput) and a custom driving profile for their estimation, which highlighted an expected battery lifetime of 10 years/ 150 000 miles.

Hoke et al., have analyzed in [10] the possibility of extending the lifetime of Li-ion batteries by following intelligent charging approaches; the authors have concluded that the lifetime of an EV-battery used for daily commuting could be extended by 50% by optimizing the charging procedure and reducing the time spent at high state-of-charge (SOC) [10]. Moreover, the same authors have presented in a method for minimizing the cost of EV battery charging using a simplified Li-ion battery lifetime model; the lifetime model is able to predict both the capacity and power fade of the battery [11].

The effect of capacity fade and power fade on the EV-battery retirement from operation was studied in [2]. Based on detailed simulations, the authors shown that defining the battery retirement at 70%-80% remaining capacity is inaccurate since the battery can provide sufficient range for unexpected trips. Furthermore, in this study it was shown that the capacity fade and not the power fade is the factor which decides the battery retirement from operation [2].

In the summarized studies, aspects concerning the ageing of Li-ion batteries used in EVs were studied based on more detailed [8] or simpler [11] lifetime models and using various driving cycle profiles. Nevertheless, in none of these papers, Li-ion batteries suitable for EV applications (such as NMC or NCA Li-ion chemistries) were tested under realistic operation conditions to obtain reliable battery ageing information. Thus, in this paper we close the existing literature gap by presenting results of the degradation behavior (in terms of capacity fade and internal resistance increase) of a NMC-based Li-ion battery. The results were obtained by performing laboratory ageing tests for a period of eleven months using a realistic EV mission profile.

III. EXPERIMENTAL DESCRIPTION

A. Lithium-Ion Battery

This investigation has been performed on a 28 Ah Li-ion pouch battery cell, which was developed in the framework of the Batteries2020 project [12]. The battery cell, illustrated in Fig. 1, is based on nickel manganese cobalt oxide (NMC) and graphite as cathode and anode materials, respectively. The cell operates within the 3V – 4.2 V voltage interval and the maximum allowed charging and discharging current-rate is 1.5C, which corresponds to a value of 42 A.

B. EV Mission Profile

The realistic EV mission profile, which was applied to the NMC-based battery cells has two distinct components: the temperature and the load current profiles.

1) Temperature Profile: The batteries were tested using a temperature profile, which is characteristic to the city of Seville, Spain and is presented in Fig. 2. During the ageing test, the temperature of the battery cell was kept constant to the values specified in Fig. 2 for a month, while the driving (load current) profile was applied at the batteries' terminals.
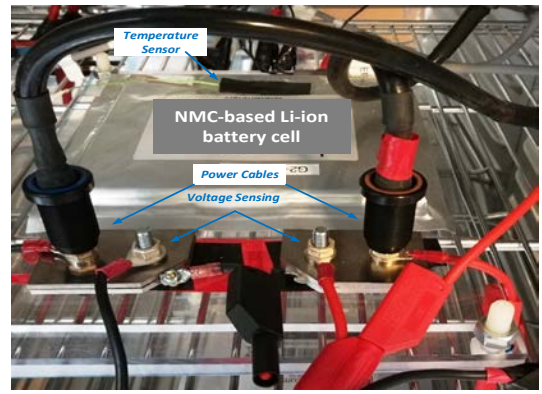

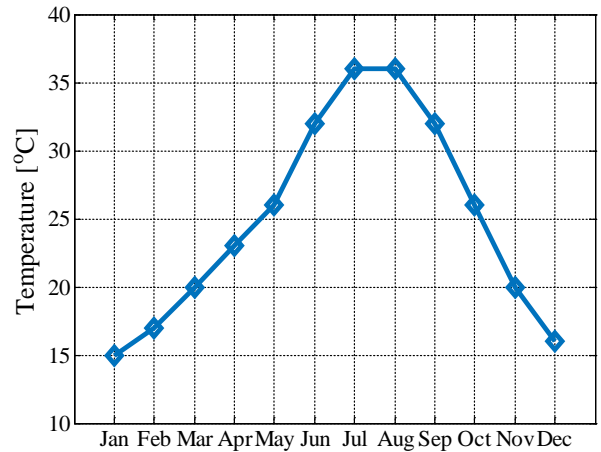Fig. 1. NMC-based Li-ion battery cell during the accelerated ageing test.

Fig. 2. Temperature profile characteristic to the city of Seville, Sapin.

2) Load Profile: For ageing the the NMC-based Li-ion cells and for determining their degradation behaviour and expected lifetime, the Worldwide harmonized Light vehicles Test Cycle (WLTC) was used. The WLTC driving cycle for a class 3 vehicle is presented in Fig. 3. This driving cycle takes 30 minutes and is composed of two sequences: the first sequence of 15 minutes simulates a period of urban driving, while the second sequence, also of 15 minutes, simulates a period of extra-urban (highway) driving.

In order to test the batteries, the driving cycle, presented in Fig. 3 (given as speed vs. time) was transformed using (1) into a current vs time profile. The obtained profile is presented in Fig. 4, where the urban and extra-urban sequences are highlighted.

$$I = \frac{\frac{1}{2} \cdot \rho \cdot S \cdot C_x \cdot v^3 + C_r \cdot m_t \cdot g \cdot v + m_t \cdot a \cdot v}{\eta \cdot V} \quad (1)$$

Where, ρ represents the air density, S represents the vehicle frontal area, C_x represents the drag coefficient, v represents the speed of the vehicle, C_r represents the rolling coefficient, m_t represents the mass of the vehicle, g represents the gravity

constant, η represents the overall efficiency of the motor, controller and converter, I represents the battery current and V represents the battery voltage.
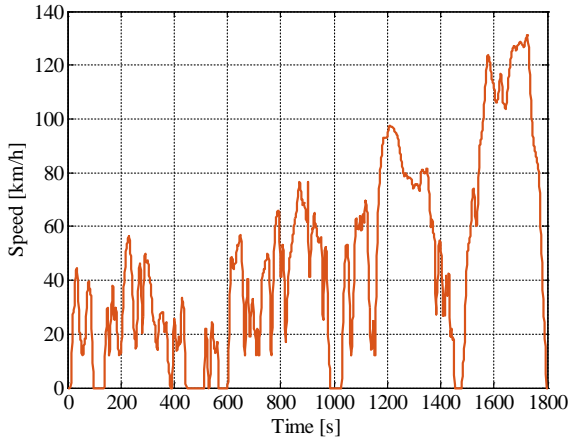

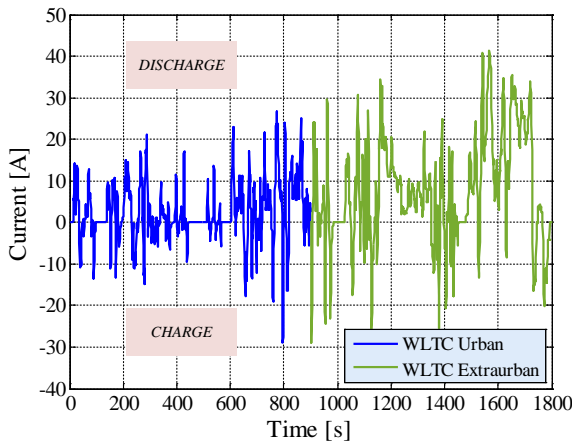Fig. 3. WLTC driving cycle for a Class 3 vehicle.

Fig. 4. Battery current profile corresponding to the driving cycle presented in Fig. 3.

The ageing test was performed on two identical NMC-based Li-ion battery cells according to the following sequence, which emulates the daily routine of an EV:

1. The battery is initially charged to 90% SOC.
2. The load profile presented in Fig. 4 is applied to the battery (i.e., the user drives the EV from home to work). During this operation, the SOC of the battery decreases to approximately 65%.
3. The battery is re-charged to 90% SOC with C/3 (i.e., the EV is re-charged).
4. The battery is in stand-by at 90% SOC (i.e., the EV waits for the next event).
5. The load profile presented in Fig. 4 is applied to the battery (i.e., the user drives from work to home the EV). During this operation, the SOC of the battery decreases to approximately 65%.

6. The battery is re-charged to 90% SOC with C/3 (i.e., the EV is re-charged for the following day).

The one-day current profile and the resulting battery SOC profile corresponding to the summarized sequence are presented in Fig. 5. By analyzing these profiles it can be observed that the Li-ion battery is operating (i.e., drive cycle and re-charging) for approximatively four hours, while for the remaining twenty hours it is on stand-by at 90% SOC. Since the cycling operation is quite limited in comparison to the stand-by operation, it was not expected that the load profile will have a high impact on the ageing of the tested NMC-based Li-ion cells. Consequently, we have decided to increase the cycling operation periods and subsequently reduce the stand-by operation periods. Thus, the two NMC-based cells were aged in accelerated manner by applying a daily ageing profile composed on approximately 22 hours of cycling and 2 hours of stand-by; the used accelerated ageing mission profile is presented in Fig. 6.
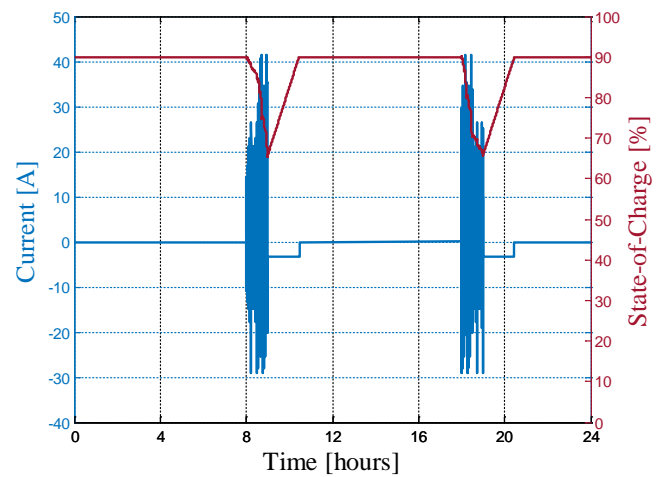

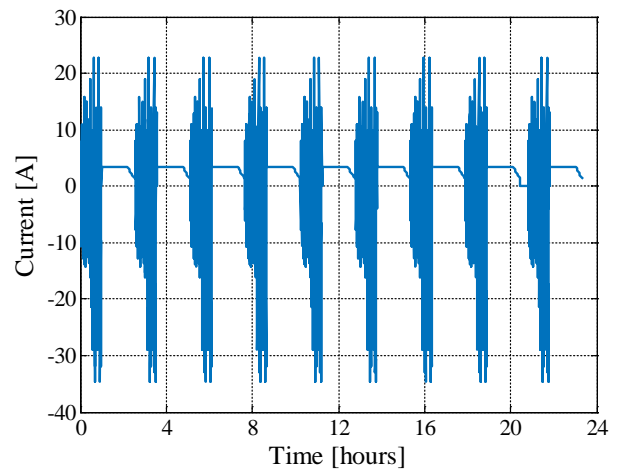Fig. 5. One-day load current profile based on WLTC driving cycle.

Fig. 6. One-day load current profile for accelerated ageing of the NMC-based cells.

IV. RESULTS

A. Voltage Profile

The voltage profile presented in Fig. 7 was measured at the terminal of one of the tested NMC-based Li-ion cells and represents the response of the battery to the one-day current profile illustrated in Fig. 6.
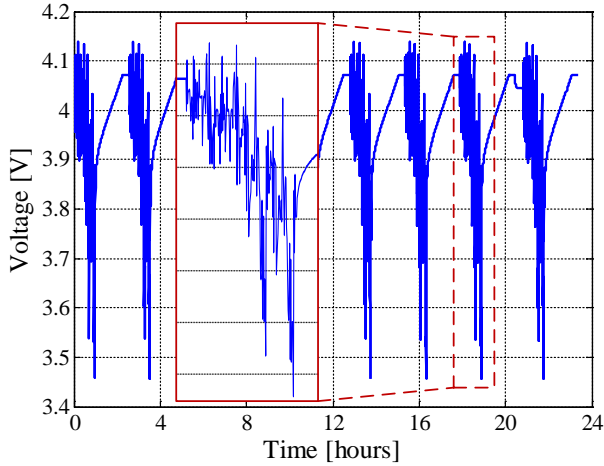

Fig. 7. Measured battery voltage profile during one day of accelerated ageing.

B. Check-up Description

The two NMC-based battery cells were aged for a period of eleven months (i.e., 356 days) using the load profile presented in Fig. 6. Furthermore, the temperature during the accelerated ageing tests was changed monthly following the profile presented in Fig. 2; the first month of test was considered August.

After each month of accelerated ageing, the two NMC-based Li-ion cells were subjected to a check-up procedure, where the capacity and the internal resistance were measured at a temperature of 25°C. The capacity of the battery was measured with C/3-rate, which corresponds to a current of 9.33 A, during both charging and discharging. In order to ensure reliable and consistent results, the capacity of the cell was measured for three times and the last recorded value was used as a reference for the ageing analysis. Furthermore, the internal resistance of the cells was measured at three state-of-charge (SOC) levels (i.e., 80%, 50%, and 20%) by applying charging and discharging current pulses of different amplitudes (i.e., 0.5C-, 1C-, and 1.5C-rate). An exemplification of the current, voltage, and temperature signals measured during the check-up are illustrated in Fig.8.

C. Capacity Fade

Fig. 9 presents the measured capacity fade of the two tested NMC-based Li-ion cells. As it can be observed, during the eleven months of accelerated ageing, the cells lost approximately 10% of their capacity. Furthermore, the same degradation behavior was obtained for the two cells, with a single exception – after the second month of testing, which is believed to be a measurement artefact.

As expected, the capacity fade has a tendency to slow down as the ageing of the battery evolves, which is a characteristic of Li-ion batteries as shown in [5], [6]. Furthermore, this behavior is also emphasized in Fig. 10, where the measured capacity fade of the tested battery cells is approximated using the power law function given in (2). The obtained capacity fade estimation function is close to a square-root-of-time function (i.e., power law function with). According to [13] and [14], the degradation of a Li-ion battery following a square-root of time function is mainly caused by the irreversible capacity loss due to the formation and growth of the solid electrolyte interface (SEI) layer.
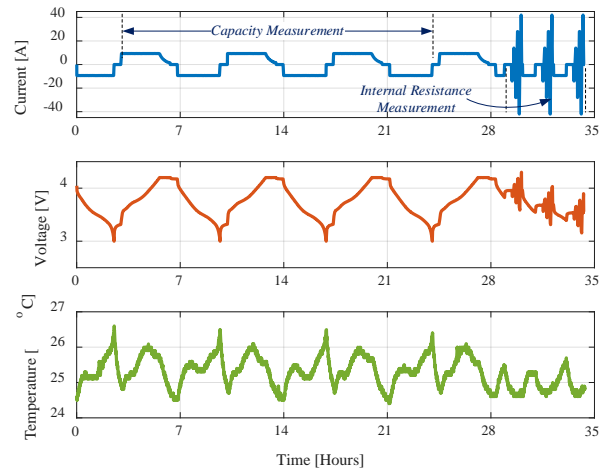

Fig. 8. Current (top), voltage (middle), and temperature (bottom) measured on the NMC-based battery cell during one the check-up procedures.
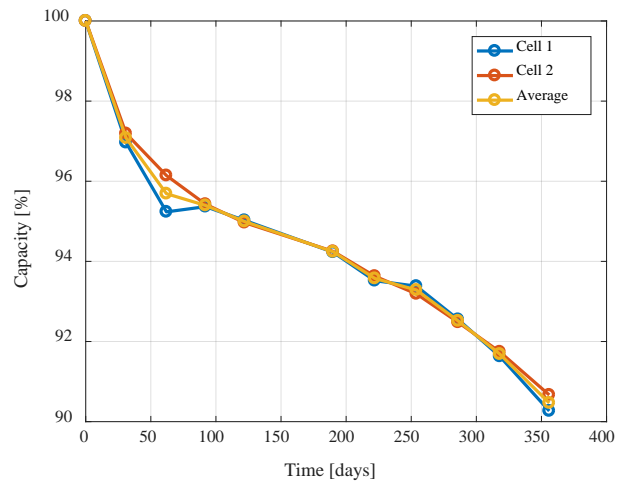

Fig. 9. Capacity fade of the tested NMC-based cells during eleven months of accelerated ageing tests.
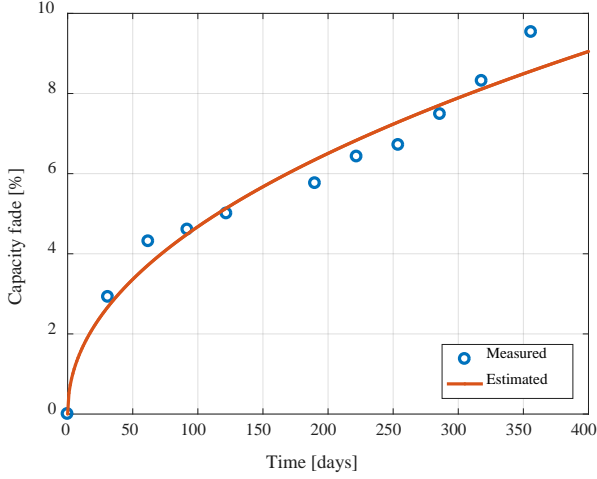
Fig. 10. Measured and estimated capacity fade of the NMC-based Li-ion cells.

Fig. 11. The correlation between the monthly measured battery capacity fade and the considered temperature.

$$C_{\text{fade}}(t)\ [\%]= 0.5221 \cdot t^{0.4761} \qquad (2)$$

Where C_{fade} represents the capacity fade of the battery and t represents the battery aging time.

Based on the capacity fade estimation given in (2) and assuming that the same mission profile will be applied, the tested Li-ion battery cells will reach an end-of-life (EOL) criterion of 20% capacity fade after approximately 5.8 years.

The relationship between the monthly measured capacity fade and the temperature during ageing is presented in Fig. 11. The highest capacity fade, approximately 3%, was measured in the first month of ageing, when the NMC-based battery cells were tested at 36°C, which represents the highest value of the considered temperature profile. On the contrary, the least degradation was obtained, when the battery cells were tested at 26°C.

The NMC-based Li-ion batteries were subjected to a total of 38685.85 Ah-throughput during the eleven months of accelerated ageing tests. This value of Ah-throughput corresponds to approximately 691 full equivalent cycles. Considering, the previous estimation of a battery lifetime of 5.8 years, we can conclude that the tested battery is able to withstand approximately 4370 cycles, which is well above the targeted battery lifetime in full EV applications.

D. Internal Resistance Increase

The power capability of the batteries is dependent on their internal resistance [15]. Subsequently, the increase of the internal resistance, due to ageing, will cause the decrease of the power capability. Thus, the evolution of the tested NMC-based Li-ion battery cells during the considered ageing tests was investigated. During the check-ups, the internal resistance of the cells was measured using both charging and discharging current pulses of different amplitudes (i.e., 0.5C-, 1C-, and 1.5C-rate) at three SOC levels (i.e., 80%, 50%, and 20%).
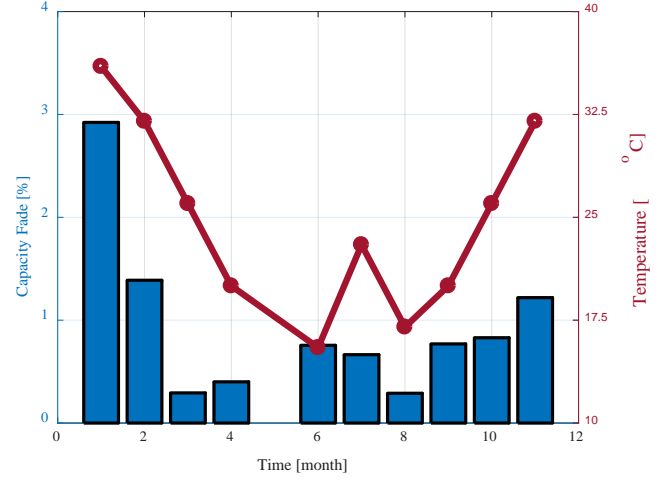
Nevertheless, further on only the results obtained for 1.5C-rate charging and discharging pulse will be presented and analyzed.

The evolution during the ageing process of the internal resistance, which was measured at 20%, 50%, and 80% SOC, is presented in Fig. 12 and Fig. 13 for the charging and discharging current pulses, respectively. Similar to the case of the capacity, results for both cells as well as the average between the two cells are presented. As it can be noticed, independent on the measurement condition (SOC and current direction), the internal resistance of Cell 2 increases slower than the one of Cell 1; this fact might have been caused by the usual manufacturing tolerances. Furthermore, the same internal resistance increase (the average value between the two cells) of 42-44% was obtained for all the three considered SOC levels, as shown in both Fig. 14 and Fig. 15.

By analyzing the results presented in Fig. 14 and Fig. 15, it can be observed that during the eleven months of testing the trend of internal resistance increase accelerates as the ageing process evolves. In order to quantify the obtained results, the trends of the internal resistance increase were fitted using a power law function. The results of the curve fitting processes are given in (3) and (4) for the charging and discharging case, respectively. According to the lifetime models (3) and (4), the internal resistance of the battery cell will increase by 300% after 3.05 years if the resistance is measured for a charging pulse and after 3.41 years if the resistance is measured for a discharging pulse. Consequently, the internal resistance and not the capacity might be the performance parameter which limits the lifetime of the tested NMC-based Li-ion battery cells (5.8 years lifetime based on capacity fade estimation against 3.05 – 3.41 years lifetime based on internal resistance increase estimation).
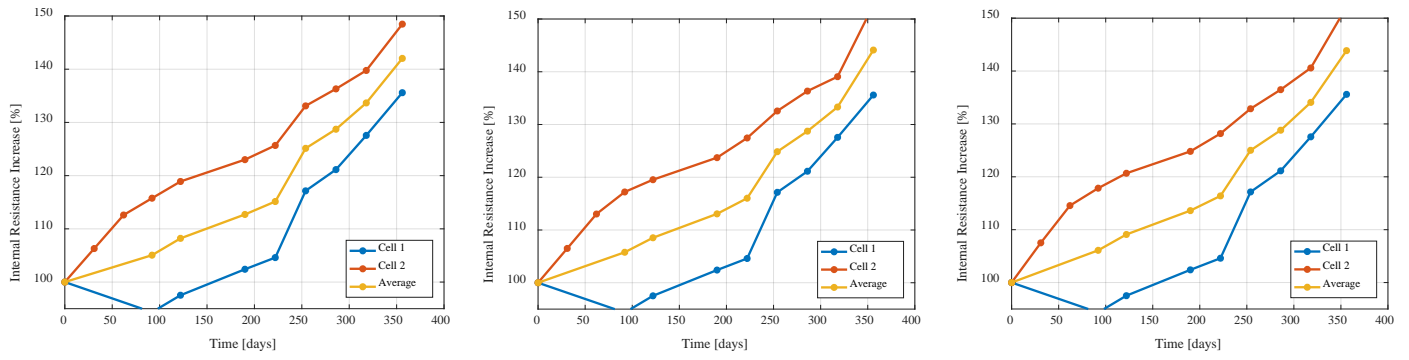
Fig. 12. Internal resistance increase measured at 20% SOC (left), 50% SOC (centre), and 80% SOC (right) for a 1.5 C-rate charging pulse.
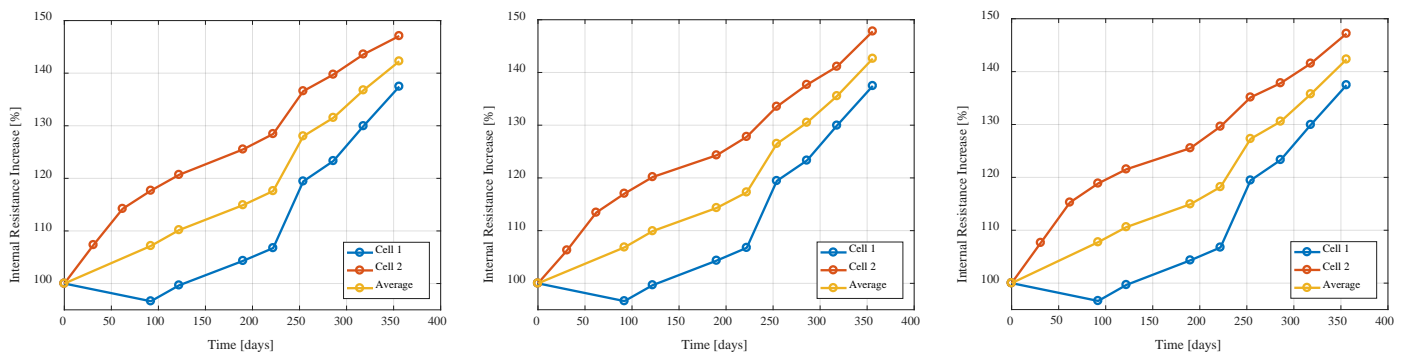

Fig. 13. Internal resistance increase measured at 20% SOC (left), 50% SOC (centre), and 80% SOC (right) for a 1.5 C-rate discharging pulse.

Fig. 14. Measured internal resistance increse at different SOC levels and estimated resistance increase; internal resistance measured during a charging current pulse.
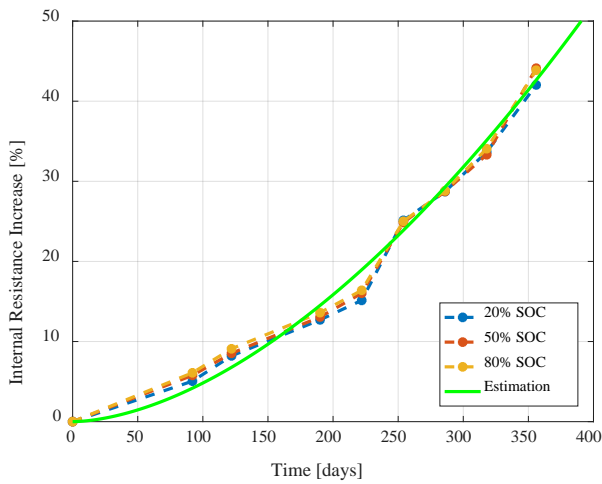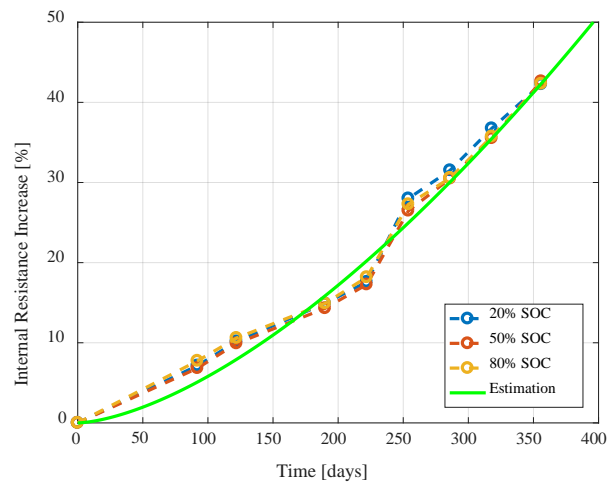

Fig. 15. Measured internal resistance increse at different SOC levels and estimated resistance increase; internal resistance measured during a discharging current pulse.

$$R_{\text{increase\_ch}}(t) \ [\%] = 0.001774 \cdot t^{1.717} \qquad (3)$$

$$R_{\text{increase\_dch}}(t) \ [\%] = 0.004328 \cdot t^{1.564} \qquad (4)$$

Where, $R_{\text{increase\_ch}}$ represents the resistance increase measuring during a charging pulse and $R_{\text{increase\_dch}}$ represents the resistance increase measuring during a discharging pulse.

V. Conclusions

In this paper we have analyzed the degradation behavior of an NMC-based Li-ion battery used in an EV application. To perform this study, the WLTC driving cycle was used; however, the battery degradation caused by a daily driving mission profile using the WLTC driving cycle is rather limited, since the battery is in stand-by for approximately 90% of time. Consequently, we have decided to accelerate the ageing process and we have applied continuously the WLTC driving cycle to the battery allowing only two hours of relaxation during each day of operation.

The results obtained during eleven months of accelerated ageing have shown a battery capacity fade on approximately 10% and an internal resistance increase of 42-44%. During the ageing period, we have observed a slowdown of the capacity fade process and a speedup of the internal resistance increase. Based on aforementioned results and trends and taking into account that during the eleven months, the battery cells were subjected to 691 full equivalent cycles, we estimated that the NMC-based battery cells would be able to withstand 4370 cycles until they will reach an EOL criterion of 20% capacity fade. However, for the tested NMC-based Li-ion battery cells the internal resistance is the performance parameter, which limits the lifetime; for the considered mission profile, a tripling of the internal resistance is obtained after approximately three years, while a 20% capacity fade is reached after 5.8 years.

Acknowledgment

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