Experimental comparison of fast-charging protocols for NMC and NCA Li-ion batteries

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***Abstract*—Fast charging of lithium-ion batteries is today one of the key technological challenges to the wide adoption of electric vehicles. In recent years several charging protocols have been proposed, having the goal of reducing the charging time without hindering the battery life. Although several among these are promising, a comprehensive comparison among them is complicated by the fact that in the existing literature each has been tested with different cells and in specific conditions. This paper provides an overview as well as an experimental comparison of such protocols on two chemistries (NMC and NCA). It is found that out of the tested protocols, only the boost charging protocol reduces charging time by up to 9% as compared to the standard constant-current-constant-voltage (CCCV) protocol. However, it also decreases the cycle life of the cell by up to 14%. The pulse charging protocols, on the other hand, increase the charging time by up to 12% but provide and increase in cycle life by up to 167%, as compared to the CCCV protocol.**

Keywords—li-ion batteries, fast-charging protocols

I. Introduction

Global warming is real and to meet the Paris agreement limit of 1.5°C, we need to drastically reduce our greenhouse gas emissions. More than 16% of the greenhouse gas emissions arise from the transportation sector. Hence, the decarbonization of this sector is fundamental in achieving the 1.5°C goal. Decarbonizing the transport sector involves moving away from internal combustion engine (ICE) vehicles towards electric vehicles (EVs). However, the market penetration of EVs is still limited by the change in consumer habits it requires. Consumers are used to spend about two or three minutes at the gas station to "recharge" their vehicle. As of today, EVs require a charging time of thirty minutes to several hours. To accelerate the transition of the consumer from ICE vehicles to EVs, faster charging times are necessary.

With today's Lithium-ion battery technologies, however, these are limited by the fact that high charging rates accelerate battery degradation [1], reducing EV life cycle and reliability. Research in battery chemistries aims at finding new materials that can withstand higher charging rates without incurring high degradation [2]. Another viable route for achieving shorter charging times without generating detrimental effect on the battery health is the design of adequate charging protocols. Charging protocols consist of a set of steps used to charge a Li-ion cell. Different types of protocols have been developed [1]. However, they are rarely compared to each other. Hence, in this paper, a set of comparable charging protocols are designed, tested on two cell chemistries and compared based on the charging time and cycle life they can achieve.

The remainder of this paper is structured as follows: Section II consists of an overview of the fast-charging protocols in the literature. Section III details the experimental setup and all the charging protocols used in this work followed by section IV, which consists of the experimental results. Section V involves discussions on the cell cycling results obtained as well as the EIS measurements made, and section VI concludes the paper.

II. Overview of Fast-Charging Protocols

A thorough review of fast-charging protocols for Li-ion batteries is presented in [1], where charging profiles are classified into six common categories, viz., constant current constant voltage (CC-CV), constant power constant voltage (CP-CV), multistage constant current constant voltage (MCC-CV), pulsed charging, boost charging and variable current profiles.

The CC-CV profile consists of charging the cell at a constant current (CC) until a certain voltage has been reached, and then maintain a constant voltage (CV) across the cell until the current drops to a very low value. This protocol is the most widely used and, thus, has been extensively studied for both the charging time and cell degradation that comes along with it. [3] studied the effect of the charging current and the ambient temperature on the aging of the cell. They showed that increasing the charging current beyond a certain limit did not reduce charging time significantly. Moreover, increasing the charging current led to lithium plating and the reduced the charging efficiency. Moreover, [3] shows that charging at low temperatures also leads to lithium plating.

The MCC-CV profile consists of multiple constant current steps (MCC) followed by a constant voltage (CV) step. [4] developed an MCC-CV profile with 2 CC steps and 1 CV step and tested it on an LFP cell. The first CC step consisted of a 4C charging until the cell voltage reached 3.6V, while the second CC step consisted of a 1C charge until 3.6V. The CV step was conducted for 5 minutes at 3.6V. They focused more on charging time and claimed that the charging protocol could charge the cell from 0% to 40% SoC within 6 minutes and full charge in 20 minutes. They also predicted a cycle life of more than 5000 cycles. However, [5] also investigated the effect of different charging protocols on cycle life. They compared the CC-CV, MCC-CV and CP-CV profiles on an LCO cell at two c-rates, 0.5C and 1C and concluded that capacity retention was highest for the CC-CV

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and CP-CV protocol, respectively, while it was lowest for the CP-CV and MCC-CV protocol, respectively.

Pulsed charging protocols introduce rest phases, and even negative current phases, during the constant current phase. The introduction of such phases aims to reduce the occurrence of lithium plating by preventing the anode voltage from dropping too low. [6] compared 1C and 0.5C average pulsed charging with 1C CC-CV on LCO cells and observed that the capacity retention was higher for the pulsed charging protocols. They concluded that the pulsed charging protocol maintained the cathode active material structure and also reduced charging time. [7] also compared the CC-CV protocol with the pulsed charging protocol. They first found the optimal pulse parameters and using these, they observed that at 0.5C, the pulse charging resulted in the charging time 47% lower than that from the CC-CV protocol. They observed improvements in the energy and charge efficiencies as well.

The boost charging protocol was first introduced by [8]. It involves charging the cell initially at a high voltage which results in a high current. After a certain time, the charging is shifted to the standard CC-CV protocol. If the current in the first stage is too high, a limit can be set on it, which makes this protocol a two-stage CC-CV protocol. [8] tested the protocol and concluded that more than 50% of the cell can be charged in 10 minutes while 30% can be charged in 5 minutes. Moreover, they claimed that this protocol did not have a significant impact on the cycle life of the cell.

Finally, a set of charging protocols in which the current profile depends on various parameters have been proposed. For example, [9] developed a constant temperature constant voltage protocol, wherein, during the first part of the protocol, the cell was kept at a constant temperature by regulating the current. [10] developed a charging protocol based on the expansion of a pouch cell due to lithium plating. The protocol was optimized such that the expansion of the pouch cell could be limited by limiting the maximum current that could be injected into the cell. [11] the sinusoidal ripple current strategy where the cell was charged with a sinusoidal current, as opposed to dc, at a frequency such that the internal resistance would be low. They demonstrated that this strategy allowed for faster charging and lower cell degradation as compared to CC-CV and pulsed charging.

Despite the plethora of charging protocols available, they are seldom compared altogether.

III. Experimental setup

A total of 12 cylindrical cells and 6 charging protocols were tested. The cells were cycled with a PEC-ACT 0550 battery tested and kept at a constant ambient temperature of 20°C by an ESPEC-ARU 1100 climatic chamber.

The PEC- ACT 0550 battery tester is equipped with 20 parallel, 5V-50A channels. Each channel can be programmed and used independently by the user. The machine has 100 μsec based internal sampling, control and capacity calculations and the FPGA hardware controls both current and voltage with a ± 0.03% FSD and ± 0.005% FSD accuracy on the respective readings.

The ESPEC-ARU 1100 climatic chamber, used to maintain the temperature at which the cells were cycled, has a volume of 1100 L and a temperature range of -45°C/180°C. Its temperature change rate is between 4K/minute and 18/K minute.

The fast-charging experiments were conducted on two sets of six cells each. The first set consisted of LG Chem Lithium Nickel-Manganese-Cobalt oxide (NMC) cells while the second set consisted of Samsung SDI Lithium Nickel-Cobalt-Aluminum oxide (NCA) cells. The electrical parameters of each type of cell are given in Table 1.

Electrical parameter	LG Chem	Samsung SDI
Cell chemistry	NMC	NCA
Form factor	2170	2170
Standard discharge capacity	4.85 Ah	4.9 Ah
Nominal voltage	3.63 V	3.6 V
Standard maximum charging voltage	4.2 V	4.2 V
Standard charging current	1.6 A	2.450 A
Standard cut-off current	242.5 mA	245 mA
Standard discharging current	1.6 A	4.9 A
Standard discharge cut-off voltage	2.85 V	2.5V
Cycle life (cycling with standard conditions up to 80% rated discharge capacity)	500 cycles	500 cycles
Maximum charging current	3.395 A (25-50°C)	4.9 A
Discharge cut-off voltage	2.5 V	2.5 V

Table 1: Electrical properties of each cell tested

Six different protocols were tested, one per cell in each set. The tested protocols were:

- Constant current constant voltage (CCCV, served as benchmark)
- Pulsed charging (PC)
- Pulsed charging with negative pulses (PCN)
- Boost charging (BC)
- Constant temperature constant voltage 1 (CTCV1)
- Constant temperature constant voltage 2 (CTCV2)

These protocols were selected to be as representative as possible of the existing literature and were designed to have comparable average charging rates. The experiments consisted in repeated cycling of the cells including fast-charging with a specific protocols and periodic diagnostic cycles. The testing procedure common to all tested cells is described hereafter, while in the following sections the details of all individual fast-charging protocols are provided.

Each cell was charged with a specific protocol, rested for 10 minutes, discharged at 1C until the cell voltage reached 2.5V and rested for 20 minutes. This constituted one cycle. Every 20 cycles, a capacity check was done. During the capacity check, the cells were charged at 0.5C until the voltage hit 4.2V and then maintained at 4.2V until the current dropped to below 0.02C. The cells were then rested for 10 minutes. Thereafter, they were discharged at 0.2C until the voltage reached 2.5V, and the discharged capacity and internal resistance were recorded. This was followed by another rest phase for 20 minutes.

A. Constant current constant voltage (CCCV)

The cell for this protocol was cycled at a current of 2C until the voltage reached 4.2V. Thereafter, the cell was maintained at 4.2V until the current dropped to below 0.05C. After the charging phase, the cell was rested for 10 minutes before the discharge phase began. Figure 1 shows the current, voltage and temperature profiles during one such charge. The CCCV protocol being the one conventionally used, it has been used as a benchmark for the analysis and comparison of the following protocols.

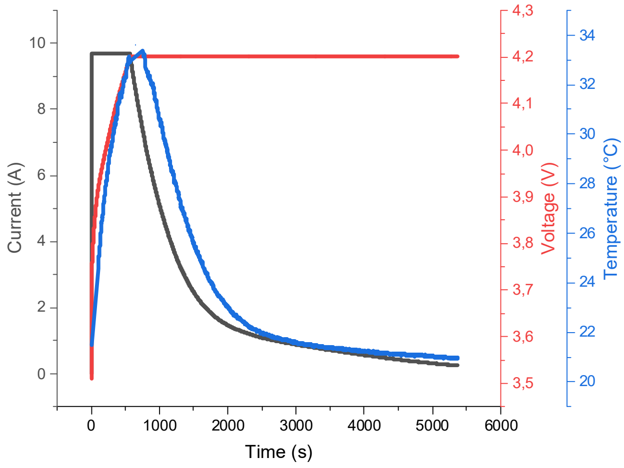

Figure 1: C-rate, voltage and temperature evolution during charging for the CCCV protocol

B. Pulsed charging (PC)

During pulsed charging, the cell was charged at 2.5C for 8 seconds and rested for 2 seconds. These values have been chosen such that the average c-rate for this protocol is the same as that of the CCCV protocol, i.e., 2C. After the cell reached 4.2V, it was continued to charge in this pulsating fashion, by decreasing the charging current to hold the cell at a maximum of 4.2V. The charging continued until the charging current reached 0.05C. Figure 2 shows a snippet of the current, voltage and temperature profiles during one such charge.

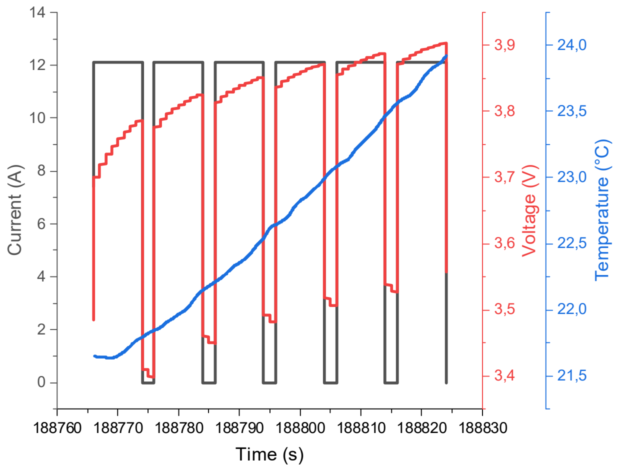

Figure 2: Snippet of the c-rate, voltage and temperature evolution during charging for the PC protocol

C. Pulsed charging with negative pulses (PCN)

This protocol consisted of charging the cell at 3C for 7.11 seconds, then discharging at 1.5C for 0.89 seconds and finally resting the cells for 2 seconds. These values have also been chosen such that the average c-rate for this protocol is consistent with that of the CCCV protocol. The voltage limit was set to 4.2V so that once the cell reached 4.2V, the charging current decreased (comparably to a pulsed CV phase). The discharge pulses continued only until the charging current was above the value of 1.5C. Thereafter, the protocol shifted to PC wherein the cell was charged at a charging current depending upon the voltage, for 8 seconds and rested for 2 seconds. The reason for this was that as the voltage increased, the charging current would drop to a level such that the charge input into the cell in the 7.11 seconds of charging would equal to that drawn out in the 0.89 seconds of discharging, effectively preventing the cell to charge further. The PC charging was continued until the charging current reached a value of 0.05C. Figure 3 shows the evolution of the current, voltage and temperature during a part of the charging phase for this protocol.

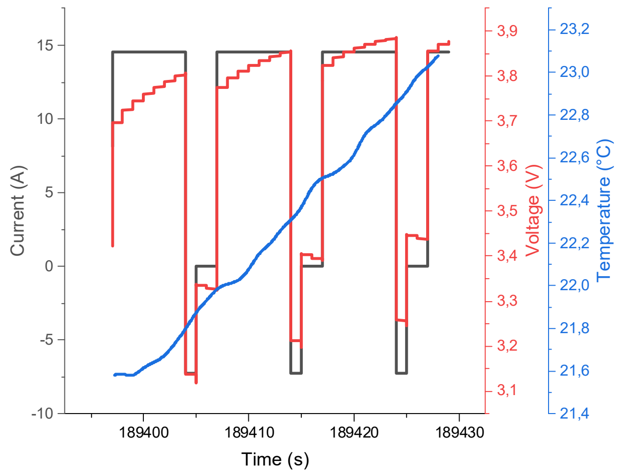

Figure 3: Snippet of the c-rate, voltage and temperature evolution during charging for the PCN protocol

D. Boost charging (BC)

In the boost charging protocol, the cell was charged at 4C until it reached 4.2V and then held at 4.2V in the first phase, called the boost period, which lasted 5 minutes. The cell was then rested for 10 seconds before it was charged at 2C until it reached 4.2V, and then held at 4.2V until the charging current dropped to below 0.05C. The evolution of the current, voltage and temperature during a charging phase are shown in Figure 4.

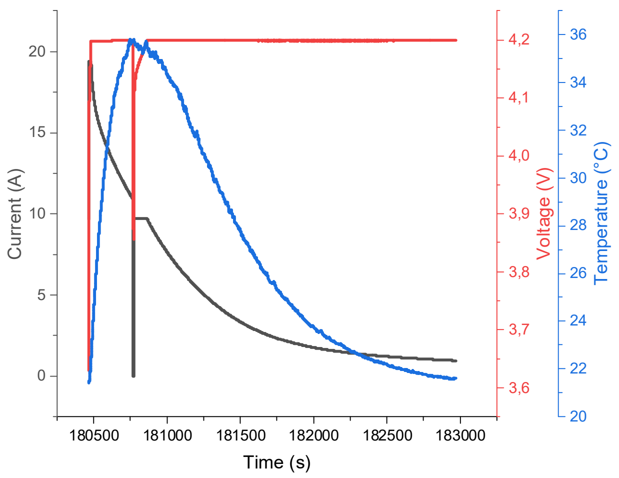

Figure 4: C-rate, voltage and temperature evolution during charging for the BC protocol

E. Constant temperature constant voltage (CTCV)

This protocol consisted of maintaining the cell surface temperature at a given value (28°C for CTCV1 and 33°C for CTCV2) by regulating the charging current. This was accomplished by monitoring the surface temperature of the cell and implementing a PID controller in the cycler. The charging started at 2C and the charging current was then regulated to, in turn, regulate the temperature of the cell. Once the cell reached 4.2V, the charging current was decreased to maintain the cell at that voltage. This continued until the charging current dropped below 0.05C. Figure 5 shows the variations in the current, voltage and temperature during the charging phase for this protocol.

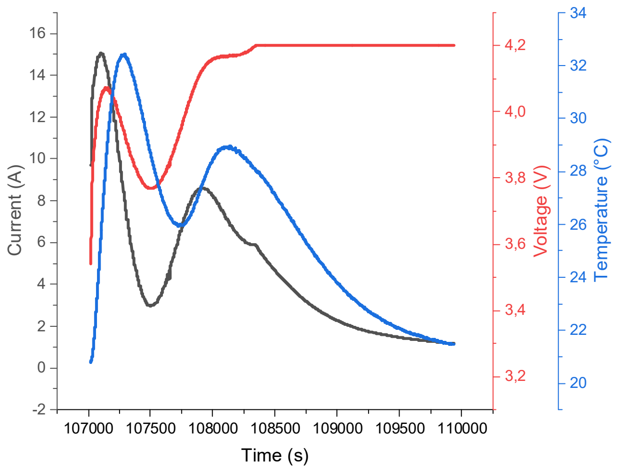

Figure 5: C-rate, voltage and temperature evolution during charging for the CTCV protocol

IV. Experimental Results

For each protocol, at every capacity check, the state of health (SoH) and state of resistance (SoR) have been assessed. SoH is given be the equation

$$SoH = \frac{C}{C_{ini}} \times 100$$

where, C is present capacity of the cell and C_{ini} the capacity of the pristine cell.

Figure 6 and Figure 7 show the evolution of the SoH with the number of full-equivalent cycles (FECs), for the NMC and NCA cells, respectively. The SoH decrease for all the tested cells has been shown below for NMC and NCA cells separately. For the NMC cells, a linear decrease in the SoH of the cells is observed, while it is non-linear for the NCA cells. Nevertheless, the cells cycled with the PCN protocol and PC protocol showed the highest and second highest capacity retention, respectively.

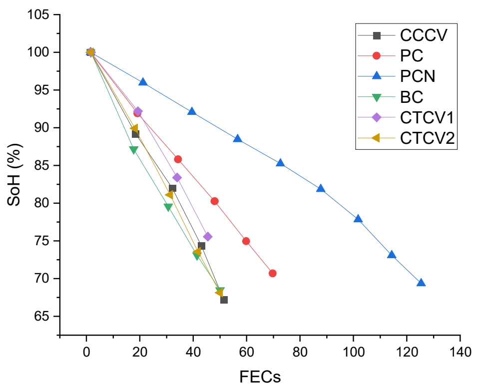

Figure 6: SoH evolution with the cycling of the NMC cells

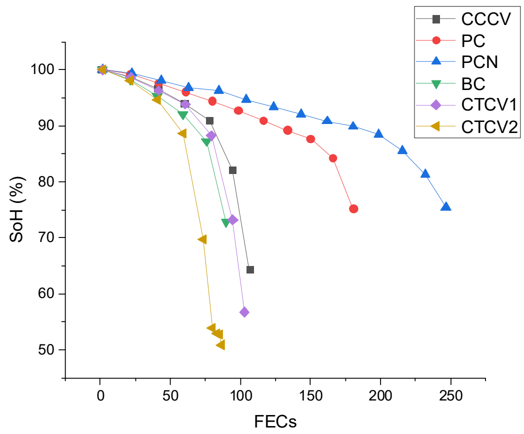

Figure 7: SoH evolution with the cycling of the NCA cells

Figure 8 and Figure 9 show the increase in the internal resistance with the FECs. The cell resistance R was calculated as the ratio of the instantaneous voltage rise or drop to the current step applied at the beginning and end of each capacity check and were calculated at 0% SoC and 100% SoC. As for SoH, the SoR is defined as

$$SoR = \frac{R}{R_{ini}} \times 100$$

where R_{ini} is the resistance value evaluated for the pristine cell.

Figure 8 and Figure 9 show the SoR recorded at 100% SoC for the NMC and NCA cells, respectively, cycled with different protocols. Akin to the capacity retention, the rise in internal resistance is linear for the NMC cells while it is non-linear for the NCA cells. This can be attributed to the difference in the chemistries of the cells. The lowest rise in resistance is recorded in the cells cycled with the PCN protocol followed by the cells cycled with the PC protocol.

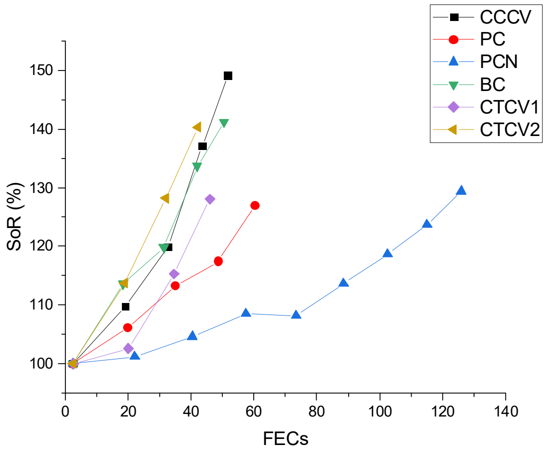

Figure 8: SoR evolution with the cycling of the NMC cells

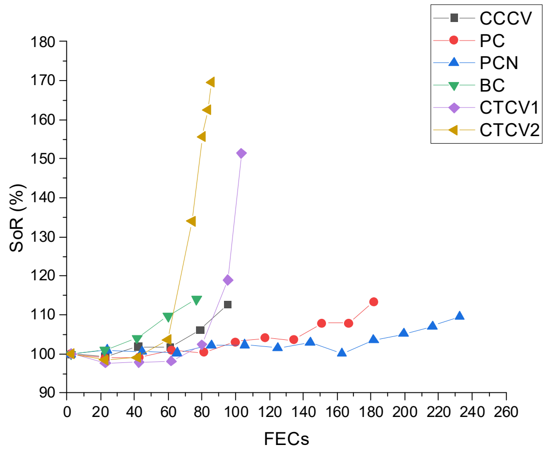

Figure 9: SoR evolution with the cycling of the NCA cells

V. Analysis and Discussion

A. Capacity retention and charging time

Charging time and capacity retention are two competing metrics. If the cell is fast charged, it is may be subject to faster degradation, while slow charging results in a longer lifetime of the cell. To analyze the effect of the different charging protocols on these metrics, the degradation vs charging time was plotted for each protocol.

The degradation rate for the different cells was extracted from the capacity loss. It was computed as the slope of the SoH vs cycle number curves which resulted in the percentage degradation per cycle. For the NCA cells which had a non-linear capacity loss trend, only the linear part of the curve was considered to compute the degradation rate. The charging time for the 1st cycle was considered. Finally, these values were normalized to the values for the CCCV protocol and plotted with charging time on the abscissa and degradation rate on the ordinate as shown in Figure 10. For reference, the charging time and degradation rate was 90 mins and 0.4 %/cycle, respectively for the NMC cell and 87.4 mins and 0.113 %/cycle, respectively for the NCA cell in the CCCV protocol.

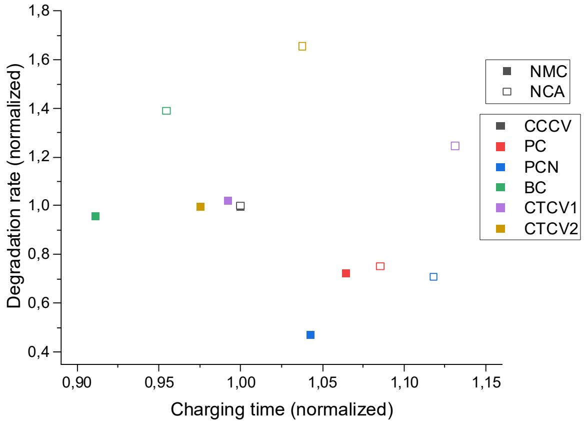

Figure 10: Degradation rate and charging time for different charging protocols, normalized to the values for the CCCV protocol for the NMC and NCA cells

For the NMC cells, the CTCV protocol values are close to those of the CCCV protocol. The BC protocol provides about 9% drop in charging time as compared to the CCCV protocol while showing similar degradation rates. The PC and PCN protocols show a 30% and 50% drop in degradation rate respectively and around 6% and 4% increase in charging time.

For the NCA cells, the CTCV protocols lie to the top right of the CCCV benchmark. The BC protocol exhibits a 5% drop in charging time while showing a 40% increase in the degradation rate. The PC and PCN protocols show a 30% and 40% gain in capacity retention respectively while gaining 7% and 13% in charging time.

For all experiments, we evaluated as well the number of FECs until end-of-life (EOL) of a cell. EOL is defined here as the point where the cell reaches 80% SoH. This metric also takes into account the non-linearities in the degradation rate of NCA cells. The normalized FECS until EOL and normalized charging times are plotted in Figure 11. This

Charging protocol	NMC			NCA		
	Degradation rate	Charging time	FECs until 80% SoH	Degradation rate	Charging time	FECs until 80% SoH
CCCV	-	-	-	-	-	-
PC	-27.81	+6.45	+44.50	-24.91	+8.54	+80.42
PCN	-53.03	+4.25	+167.76	-29.05	+11.79	+145.29
BC	-4.02	-8.86	-7.19	+39.07	-4.53	-13.64
CTCV1	+2.22	-0.80	+17.04	+24.59	+13.12	-8.60
CTCV2	-0.25	-2.48	-2.48	+65.66	+3.78	-31.52

Table 2: Percentage increase/decrease in parameters with change in charging protocol (CCCV protocol as basis)

metric shows an increase in number of FECs until 80% SoH of about 45% and 170% for the PC and PCN protocols, respectively for the NMC cells while it shows an 80% and 145% increase for the NCA cells. For reference, the charging time and FECs until EOL was 90 mins and 33 cycles, respectively for the NMC cell and 87.4 mins and about 96 cycles, respectively for the NCA cell in the CCCV protocol. The percentage increase/decrease in the various metrics for the different charging protocols with the CCCV protocol as a basis, has been given in Table 2.

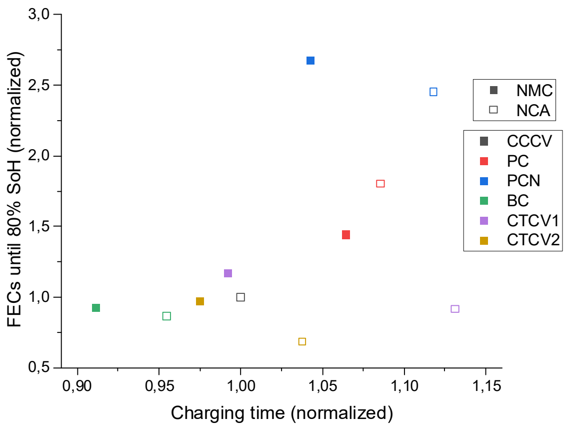

Figure 11: FECs completed at 80% SoH and charging time for different charging protocols, normalized to the values for the CCCV protocol for the NMC and NCA cells

The analysis above shows that the PCN and PC protocols provide considerably longer lifetime for charging time values comparable to those of the other tested protocols. The low degradation rate for the PCN and PC protocols could be explained by the low average cell voltage during charging. At high voltages, the anode potential vs Li+/Li drops below 0 V, and this leads to lithium plating on the anode, causing capacity loss [3]. The average voltage for each of the protocols over the first 20 charging cycles is shown for the NMC and NCA cells in Figure 12. For the PC and PCN protocols, since they are dynamic protocols, the cell voltage changes in the charging pulse, discharging pulse and rest period during the charging phase. This can be seen in Figure 2 and Figure 3. Hence, for the calculation of the average cell voltage during charging, only the voltage during the charging pulse is considered. The voltage during the rest period and the negative pulse that occur while charging has not been considered since their effect is already included into the voltage during the charging pulse. The average voltage during charging is lowest for the PCN and PC protocols, possibly avoiding the degradation/stresses that occur at high voltages.

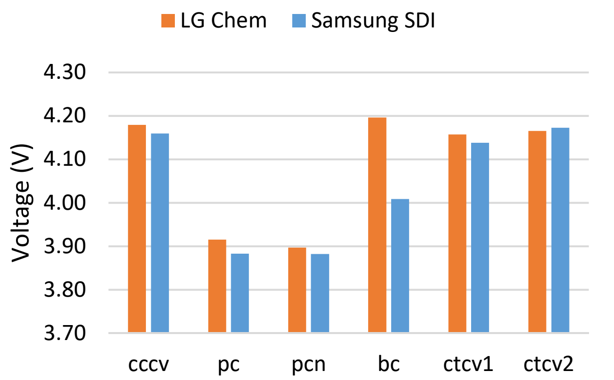

Figure 12: Average cell voltage during charging for 1st 20 cycles for various charging protocols

VI. Conclusion

The impact of different charging protocols on the degradation and charging times of the cells has been investigated. Five different types of charging protocols were investigated on two sets of cells of different chemistries (NMC and NCA). The CC-CV protocol is a standard and was thus considered as a basis to evaluate the other protocols.

Out of all the protocols, only the BC protocol showed a reduction in charging time in both the cells (approx. 9% for NMC and 5% for NCA). However, this reduction came with a reduction in the FECs until 80% SoH as well. The CTCV protocols did show a reduction in charging time for the NMC cells but did not do so for the NCA cells. Finally, the PC and PCN protocols displayed a slight increase in charging time compared to the increase in the FECs until 80% SoH. This increase in the number of cycles was attributed to the low average voltage that the cell experiences while using these protocols.

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