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Adaptive Fast Charging Strategies

How to optimize fast charging strategies to ensure longer durability of batteries

Master's thesis in Sustainable Energy Systems

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MASTER'S THESIS

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An investigation of how to optimize fast charging strategies to ensure longer durability of batteries

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Abstract

This experiment aims to test a charging procedure that can improve the durability of battery cells, that is to ensure longer lifetime of the cells. To avoid variations in the results the same battery chemistry is used in all tests which is a Li-ion battery with a Li-NMC cathode and a graphite anode. Surface temperature of the cells is kept at 35 °C to avoid any variations between tests. Limiting factors are the number of cells being tested, the type of cells being tested (only commercial pouch cells) and that the cells are only being cycled for about 3 months. The first fast charging protocol tested (FCP 1) has a current which is specified for each SOC level and does not change in between cycles; hence all cycles are run with the same current values for each SOC level. The second fast charging protocol (FCP 2) has a constant C-rate, but the current is adjusted according to the capacity after every 50 cycles.

The results are evaluated with several different measurement parameters, capacity retention, charging time, energy throughput and direct current internal resistance are all measured continuously during testing. A teardown is performed and scanning electron microscope (SEM) is used to observe any differences in degradation effects. When fast charging according to the first protocol with a constant current throughout the cycling, the capacity retention is between 58-62%. However, when adjusting the current based on battery capacity, the capacity retention is 85-86%. Similar values and trends are seen for energy throughput. Charging time decreases with 33-48% for the first protocol while staying almost constant when fast charging with the second protocol, only decreasing with 2-8%. The SEM shows severe degrading on cell 2, fast charged according to FCP 1, strengthening the results. The SEM gave indications of degraded electrolyte as well as possible lithium plating.

Tests on three electrode PAT-cells are also performed and evaluated, however due to issues with the equipment, the PAT-cells are not cycled for the full duration as planned. The experiments on commercial battery cells show clear indications that adjusting the current based on battery capacity increases battery lifetime while keeping charging time relatively constant during cycling. Future works on how to improve the testing method and increase the reliability of the results are discussed and suggestions on other tests are presented at the end.

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List of Abbreviations

<i>CO₂</i>	Carbon Dioxide
<i>ICE</i>	Internal Combustion Engine
<i>EV</i>	Electric Vehicle
<i>SDG</i>	Sustainable Development Goals
<i>CHP</i>	Combined Heat and Power
<i>BEV</i>	Battery Electric Vehicle
<i>PHEV</i>	Plug-in Hybrid Electric Vehicle
<i>FC</i>	Fast-Charge
<i>FCP</i>	Fast Charging Protocol
<i>RPT</i>	Reference Performance Test
<i>EOL</i>	End of Life
<i>BOL</i>	Beginning of Life
<i>Wh</i>	Watt-hours
<i>LiPF₆</i>	Lithium Hexafluorophosphate
<i>TiS₂</i>	Titanium Disulfide
<i>V₂O₅</i>	Vanadium Pentoxide
<i>PE</i>	Polyethylene
<i>Li</i>	Lithium
<i>LiCoO₂</i>	Lithium Cobalt Oxide
<i>LiMO₂</i>	Lithium Oxides where M is either Manganese, Cobalt or Nickel
<i>NMC</i>	Nickel Manganese Cobalt Oxide
<i>Li-NMC (LiNi_{1-y-z}Mn_yCo_zO₂)</i>	Lithium Nickel Manganese Cobalt Oxide
<i>Li-ion</i>	Lithium ion
<i>LTO (Li₄Ti₅O₁₂)</i>	Lithium Titanate
<i>SEI</i>	Solid Electrolyte Interphase
<i>C-rate</i>	Charge rate
<i>SOH</i>	State of Health
<i>CC</i>	Constant Current
<i>CV</i>	Constant Voltage
<i>CC-CV</i>	Constant Current Constant Voltage
<i>CP-CV</i>	Constant Pressure Constant Voltage
<i>MCC-CV</i>	Multi-stage Constant Current Constant Voltage
<i>VCP</i>	Variable Constant Profile
<i>SOC</i>	State of Charge
<i>NO_x</i>	Nitrogen Oxides
<i>SO_x</i>	Sulphur Oxides
<i>UNDP</i>	United Nations Development Program
<i>EB</i>	Electrical Bus
<i>BMS</i>	Battery Management System
<i>SEM</i>	Scanning Electron Microscopy
<i>BSE</i>	Backscatter Electrons
<i>DCIR</i>	Direct Current Internal Resistance

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1. Introduction

One of the biggest societal concerns for our and future generations is the progressive modification of Earth's climate, usually identified as "climate change". This phenomenon has an anthropic nature, resulting from an extensive use of fossil fuels. When burning fossil fuels, greenhouse gases such as carbon dioxide (CO₂), are being emitted to the atmosphere, affecting the climate and environment [1].

Many new innovations and technology-upgrades that have been adopted during the past few decades (such as improved electrical vehicles (EV) and renewable energy technologies), have had the initial goal to reduce the impact on the environment [2]. The energy sector has been based on fossil fuels for a long time where coal, natural gas and oil are the most used resources. These resources emit a lot of CO₂-gases when being combusted and used for production of heat and electricity [1].

A transition in the energy system sector towards cleaner energy alternatives started during the beginning of the 21st century and has led to an increase in renewable resources and less use of fossil fuels. The same started to happen within the transportation sector and the transition from fossil fuels to other alternatives begun, from vehicles with internal combustion engines (ICE) to EV [3]. The conventional ICE-vehicles that dominated human streets from the 20th century have now started to develop into something new. EVs have been on the market for a long time but have seen an enormous breakthrough during the past 15 years [4]. The fundamental technology of EVs is to charge the battery of the car with electricity, then converting the electrical energy to mechanical energy which is used to drive the car and thereby substitutes the fuel [5].

Since the environmental issue has been one of the main topics since the United Nations Conference on the Human Environment in 1972 when the Brundtland report is published, the EVs technology has been on the agenda and both positive and negative impacts have been identified. Therefore, it is critical to investigate the positive and negative impacts on the environment from EVs in order to see if, and how, EVs can support guidelines towards a sustainable development. One of these guidelines, used globally, are the United Nations Sustainable Development Goals (SDG) which are discussed further in this report. There are in total 17 SDG's where three of them are more applicable to this project. These are SDG 9; Industry, innovation and infrastructure, SDG 11; Sustainable cities and communities and SDG 13; Climate action. There is little doubt that the implementation of EVs will impact both society as well as the environment [6].

With an increased amount of EVs, the demand for auxiliary services is steadily increasing and one of these services is fast-charging stations to be able to charge the EV fully in less than one hour [7].

The consumers need for fast charging (FC) is increasing noticeably. Fast charging is mainly used for commercial cars when driving long distances where the option of waiting 4-7 hour for the car to be fully charged is not an option, for example vehicles which do not have the option for long periods of rest such as taxis or trucks where it is crucial to keep the vehicle moving at all times. It can also be of use in densely populated areas with restricted access to personal charging stations.

The process of fast charging an EV entails the use of high charging currents to be able to reach about 80% of the maximum battery level in around 30 minutes. Repeated passage of high

currents is damaging to the battery and will deteriorate it quicker than with charging longer times with lower current. Since an EV battery has an end-of-life (EOL) of around 70-80% of its original capacity [8] it is crucial to either limit the number of fast-charge events or to come up with new ways of fast-charging which keeps battery degradation to a minimum.

The main goal of our project is to find optimal fast-charging strategies which combine short charging times while preserving battery life and to discuss on how to best implement these strategies into current systems.

1.1 Aim and scope

The aim of this thesis is to investigate how to further improve an EV fast-charging strategy while at the same time making sure not to compromise the durability and safety of their batteries. A fast-charging strategy is suggested and evaluated by performing tests on battery cells at different scales. The same tests are performed on both larger (commercial cells) and smaller (PAT-cells) scale.

The main subgoals are:

- Study different charging rates during fast charging and the effects they have on battery lifetime.
- Better understanding of what factors (for example current or voltage) affect the degradation.
- Trying to optimize and improve current fast charging protocols.
- Evaluating the level of reproducibility of the results from the commercial battery cells using the same material but in smaller cells.

2. Theoretical framework

2.1 Transformation in energy- and electricity production

As previously mentioned, a transition in the energy system towards cleaner energy started, to reduce the negative impact on the environment. Together with the negative effect on the environment, some of the resources used in fossil-based energy production are getting scarcer, such as oil and natural gas due to high short-term demands [9]. This is one of the reasons to why the shift to a clean and 'green' energy system has been on the agenda for many years [1]. However, this shift comes with other types of scarcity in resources as well, such as metals for solar PV (photo-voltaic) panels and batteries [10]. The energy-shift is not only about the environmental aspect but the economic as well as the social perspective also plays a vital role. From an economic point of view, the fossil-based fuels have for a long time been cheaper and more beneficial than renewable energy sources, like wind and solar power. Fossil-based energy sources are more beneficial since they can produce energy at all hours and are not dependent on weather conditions for their production, hence they are able to provide energy at all hours to supply the whole demand [11].

As technology developed and became cheaper, some of the renewable energy sources became more integrated into the energy system and has replaced parts of fossil-based energy sources in multiple countries. For example, in Sweden and Finland, more than 40% of the energy demand is provided by renewable energy systems [2]. The main renewable resources are hydropower, wind power, solar power, biofuels and combined heat and power (CHP) production based on waste or biomass as fuel. In some countries, such as Sweden and Norway, both hydropower and CHP have been a part of the mix since before the switch to cleaner energy globally. However, the fuel in CHP production has been switched from coal to a biofuel or waste in many production sites [12]. The biggest increase in capacity has been with wind power and solar power depending on the climate for each country. For example, in Ireland and Germany there has been a higher increase in the capacity of wind power, while a country like Spain has had a big increase in solar PV as well as wind power [2].

2.1.1 Transition in the vehicle industry

As the energy system started to change and develop, it was inevitable for a transition towards cleaner transportation within the vehicle industry. Vehicles have for a long time been based on oil as fuel, more specifically diesel and petroleum, where both lead to high emissions of CO₂ into the atmosphere as well as air pollution [3].

As societies have developed a more stable infrastructure and larger connections between highways, more vehicles have been used on an everyday basis in industrialized countries [13]. Therefore, the switch to more clean fuels in vehicles have been an urgent problem as well. Different types of biofuels have been produced and tried where some have had a greater success. They have all been based on the goal to recreate the same fuels used today but from a clean resource such as corn, rapeseed, or sugarcane. One of the main issues and concerns regarding biofuels and production of biomass is the competitiveness of the resource. The resources available today will most likely not be enough for biofuel production for the whole road transport sector [14].

In an EV, the fuel is substituted with a battery, hence the EV is driven by electricity produced by the battery. The development of EVs has progressed over a long period of time. Already back in the 1800th century, the first electric cars had been developed. However, with the

upcoming of gasoline, which has been the cheaper alternative, the development of EVs paused [5].

When GM released their first EV back in 1996, an interest in this technology started to arise. However, with the EVs being very exclusive and expensive, this technology was not able to fully compete with the conventional car. By the year 2000 Toyota released the first hybrid car, running on both electricity and gasoline, this became a success and opened the door for further development. When Tesla Inc. released their first EV back in 2006, they gained a lot of attention and support, which also inspired other manufacturers of cars to do the same [5].

As these companies and suppliers started to develop batteries and power electronics for EVs, more car developers entered the market and started to produce their own EVs as well. This led to EVs being more competitive in the vehicle-market and more integrated into society. With an increased percentage of vehicles being electric, more pressure was put on the society to adjust for these cars which includes demand for more charging stations both in urban and suburban areas.

Today, more EVs are seen in urban and cities among people with high incomes where the welfare is well established. The majority in these well-established areas has at least one car per household, either an EV or an ICE-vehicle. EVs are still a bit more expensive and are trickier to own since the charging often needs planning in a more thorough way than for a car with an ICE since charging fully takes a lot longer time than filling a tank with gasoline [15].

2.1.2 Electric Vehicles in Sweden

From a global perspective, the amount of EVs have increased rapidly during the last decade. In IEA's report from 2021, there is a clear trend where an increase in the use of battery EVs (BEVs) and plug-in hybrid EVs (PHEVs) can be seen. Looking at these two individually, there is a higher share of use of BEV than PHEV globally. In 2020, there were more than 10 million EVs on the roads across the world. The countries with the highest share of EVs is by far, China and the United States. Europe has had a rapid increase of EVs as well, in both BEVs and PHEVs. Further, in 2017, IEA started the "EV30@30" campaign which has the goal to reach 30% sales share for EVs by year 2030. This campaign aims to increase the use and deployment of EVs globally [4].

In Sweden, the highest share of EVs used in 2020, were PHEVs where there were 122 977 registered, while there were 55 734 BEVs registered which can be seen in Figure 1. The share of sales of EVs increased from 11.4% to 32.3% from 2019 to 2020. By this it can be concluded that the total amount of different types of EVs in Sweden, in 2020, were 178 711 cars [16]. According to [17], the average battery capacity of a full EV is 61.7 kW. Some manufacturers have a much higher capacity of the battery, up to 118 kWh. The same source has published the full range of EVs where the average is 322 km [18].

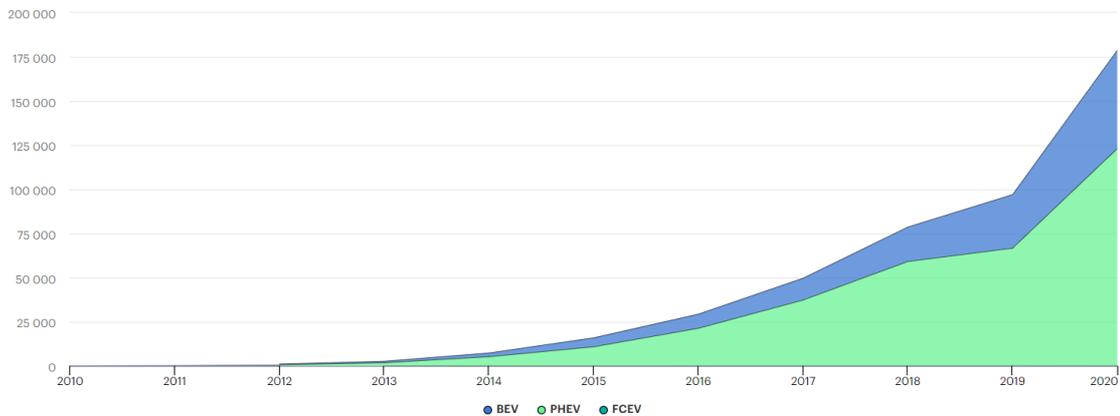


Figure 1. EV stock in Sweden 2010-2020. Figure taken from [16].

2.2 Batteries

Batteries have been proven as a very useful and applicable technology during the course of history. One of the most valuable qualities of the battery technology is that it comes in a variety of sizes and materials depending on what task it needs to fulfill. It can deliver energy instantly; it's simple to recharge and is excellent for short term storage which makes it suitable for day-to-day use [19].

A battery can come in a variety of sizes and materials but all of them are built on the basis of the electrochemical cell. The cell can store chemical energy which can be released as electrical energy when discharged, furthermore, depending on the type of battery, this process can be reversed when charging the battery [20].

A cell consists of a cathode which is the positive electrode made from materials which can donate an ion such as TiS_2 , V_2O_5 and $\text{LiNi}_{1-y-z}\text{Mn}_y\text{Co}_z\text{O}_2$ [21]. As well as an anode which is the negative electrode and should be able to accept ions from the cathode, this have mainly been graphitic carbon [22]. These are divided by a separator which is used to avoid contacts between the two electrodes which would short-circuit the cell. It's important that this material is non-conductive and is often made from some sort of polyethylene (PE) [23].

An electrolyte is a solvent in the battery that allows for transfer of ions between the separator but not electrons, where LiPF_6 is one of the most used [24]. A connector leads the electrons from the cathode through the device being supplied with electricity to the anode on the other side. These connectors are made from material such as copper and aluminum which doesn't take part in any reactions occurring in the cell [15], [25].

Figure 2 gives an overview of the standard structure of a battery cell. A load is connected between the two current collectors allowing the electrons to run from the cathode to the Anode, supplying electricity to the load.

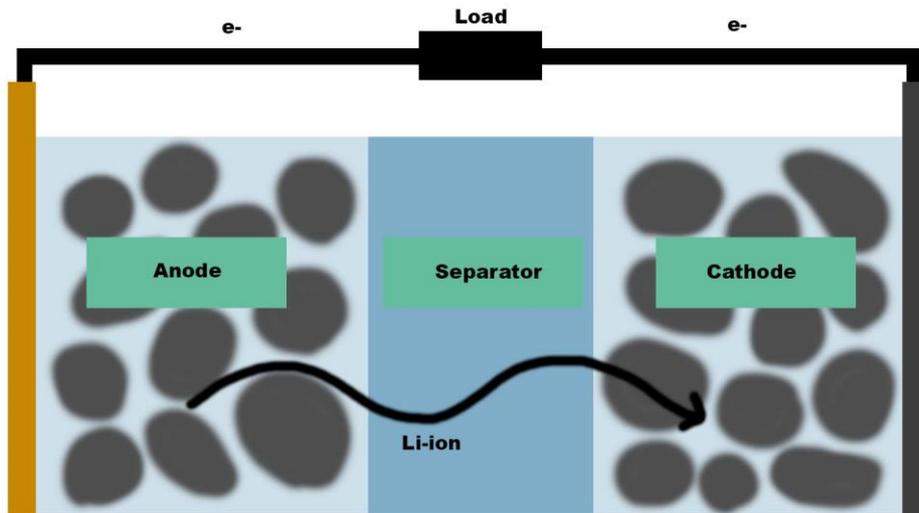


Figure 2. Simple overview of a standard battery cell. Data for the figure taken from [25].

2.2.1 Li-ion battery-cells

Li-ion batteries comes in many different varieties, from smaller coin-cells used in home appliances to larger pouch cells mounted inside modules in EVs. This experiment is focused on cells used in the latter but will also treat smaller cells created from the same material as the larger ones. Cells in EVs are mounted together to construct a battery that can deliver the desired capacity to best match the car. Different battery sizes can be achieved just from connecting different amounts of cells together. Figure 3 is presenting an example of a pouch cell; this one is experimented with at Chalmers Battery Lab.

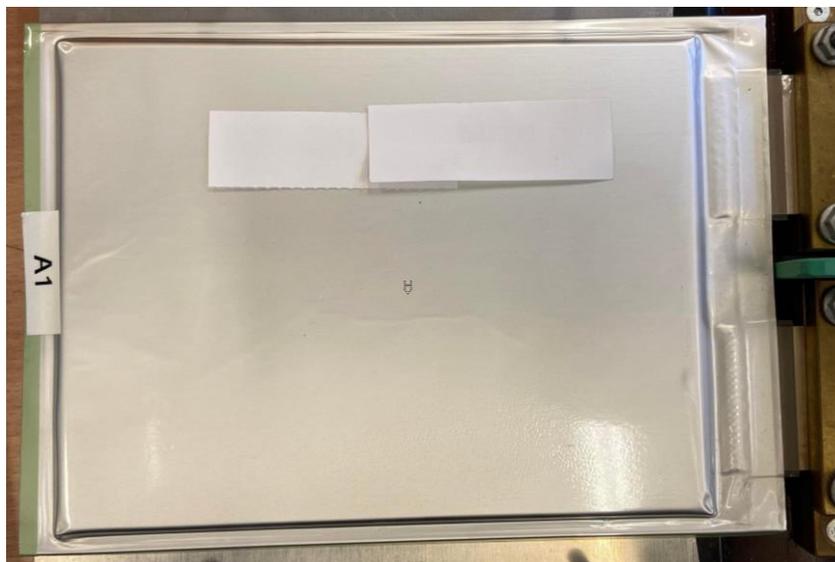


Figure 3. Example of a pouch cell. Picture is taken at the Battery Lab at Chalmers.

There are several materials that can be used for a battery cell, but Li-ion batteries have many qualities that makes them very suitable for use in the EV industry. A Li-ion battery have a high energy density, has a low self-discharge, require low maintenance and can be constructed to better match the performance of the car [26]. Li-ion batteries can use several different chemistries such as LiCoO_2 , LiMO_2 and Li-NMC ($\text{LiNi}_{1-y-z}\text{Mn}_y\text{Co}_z\text{O}_2$) [27], in this experiment

the focus is on Li-NMC (lithium nickel manganese cobalt oxides) as they are used widely in commercial cells.

2.2.2 Anode – Graphite

Graphite has been a key component in a Li-ion battery for a long time where it serves as the anode material. It contains several different properties which makes it an excellent anode. From material properties such as high energy density (Wh/m^3), high power density (W/m^3) and long cycle life to commercial and economical properties such as low cost and high availability compared to other alternatives such as LTO ($\text{Li}_4\text{Ti}_5\text{O}_{12}$) [22].

Figure 4 is presenting a graph containing the voltage profile of a graphite anode and a Li-ion cathode respectively, during charging from SOC 0 to SOC 1. The figure was used to validate that the anode material was functioning properly and had not damaged. Deviation from the curve would have meant that the cell was damaged, or the material was not correct.

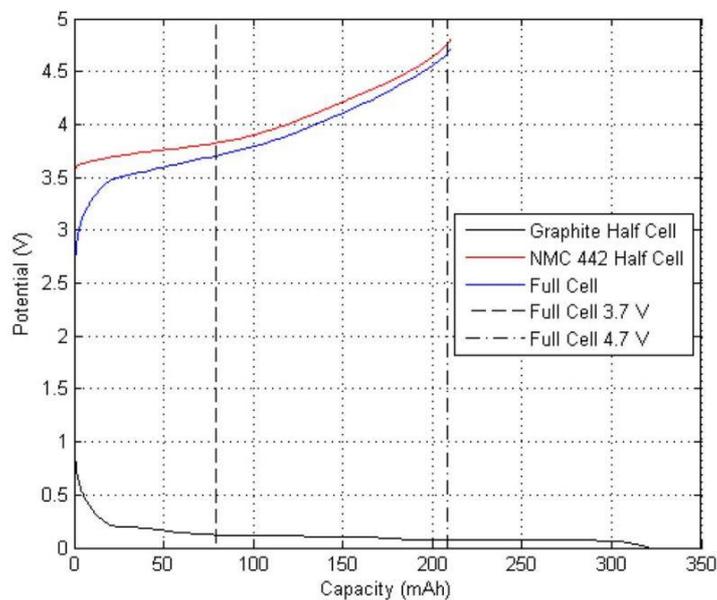


Figure 4. The voltage profile of a Li-ion NMC cathode and a graphite anode cell against specific capacity during a full charge cycle from SOC 0 to SOC 100. Dynamic cell voltage also specified by the blue line which is the difference in voltage between the cathode and anode. Figure taken from [28].

2.2.3 Cathode – Li-ion NMC

Usage of Li-ion with NMC has been very prevalent throughout several decades where several different chemical structures have been tried and used in commercial products [29]. Table 1 shows that the Li-NMC has a very high specific capacity (capacity available per g electrode) compared to other materials which is desired in an EV battery. Average potential does not impact cell performance but is rather a measurement which can be used to evaluate battery health.

Table 1. Several battery chemistries with their specific capacity and their average potential, value in parenthesis specify practical capacity. Average potential is the mean value of the voltage during cycling. Data taken from [29].

Compound	Specific Capacity (mAh g ⁻¹)	Average potential (V vs Li ⁰ /Li ⁺)
LiCoO ₂	272 (140)	4.2
LiNi _{1/3} Mn _{1/3} Co _{1/3} O ₂	272 (200)	4.0
LiMn ₂ O ₄	148 (120)	4.1
LiMn _{3/2} Ni _{1/2} O ₄	148 (120)	4.7
LiFePO ₄	170 (160)	3.45
LiFe _{1/2} Mn _{1/2} PO ₄	170 (160)	3.4/4.1

The NMC cathode has a very high specific capacity as well as voltage which leads to an energy density, much higher than other Li-ion based cathodes. This high energy capacity comes however with a tradeoff where the cell becomes a lot more sensitive to higher temperatures and a very noticeable deterioration can be seen at temperatures around 50 °C [29]. This might not be optimal in regard to fast charging but the other qualities of the NMC cathode outweighs this drawback.

Figure 4 also shows the voltage profile of a Li-ion NMC from SOC 0 to SOC 1. This was used as a reference to validate that the cathode was performing as expected. Deviation from the curve would have to mean that the cell was damaged, or the material was not correct.

2.2.4 Electrolyte

The electrolyte is another core component of a battery cell and consists of salts mixed with solvents to create a solution which acts as a mediator between the anode and cathode electrodes to allow for ions to pass through while stopping electrons [20].

A very popular electrolyte used in Li-ion battery cells is the LiPF₆ salt dissolved in ethylene carbonate and other carbonates, this is also the electrolyte used for this experiment. The solution has a few qualities which makes it an attractive choice for battery cells such as high ionic conductivity, performs well during battery operating temperatures, high durability and favors formation of the SEI [30].

2.2.5 SEI

SEI is a protective layer in the Li-ion cell which is formed between the surface of the graphite anode and the electrolyte. This layer is created by Li⁺-ions traveling from the cathode, through the electrolyte, to the anode. During this process some Li⁺-ions will be reacting with degraded products of the electrolyte, the degradation of the electrolyte is most prominent during the first charges of the cell when the anode surface is in direct contact. When the SEI is formed, the electrolyte is protected from the anode and further degradation is prevented while at the same time allowing the Li⁺-ions to freely travel through [31].

The SEI is very much affected by temperature where a low formation temperature of 30 °C or less generally leads to a thin, unstable SEI layer while a high temperature formation temperature of 60 °C or higher creates a thicker and uneven SEI layer. It's therefore very important to run a something called a formation cycle where the cell is charged at low C-rates with controlled temperature to avoid a deformed SEI layer.

The growth of the SEI is to be expected during normal operations and is hard to avoid but can be limited by restricting factors which influence the formation of the SEI. Deep discharge and temperature are two of the main factors to consider. Charging and discharging of the cell at high temperatures ($>60\text{ }^{\circ}\text{C}$) might lead to degradation of the layer which can cause a performance decrease and aging of the cell [32].

2.2.6 EL-Cells & PAT-Cells

EL-Cell is a company which produces electrochemical test equipment and test cells. The product used in this study is known as a PAT-cell which is a 3-electrode cell for Li-ion battery materials, this can be seen in Figure 5 and Figure 6. The PAT-cell is a sort of coin cell which can be directly connected to the test equipment and the software. Through the software multiple different tests can be performed by writing different programming scripts, therefore it becomes easier to build and perform tests on smaller scale. The PAT-cell consist of an insulation sleeve, an upper- and lower plunger, a screw cap, and a cell base. Among these, there is also a reference electrode which is included. Hence, everything is included in the PAT-cell except for the anode material, cathode material and electrolyte which needs to be provided externally [33].



Figure 5. Description of a PAT-cell. Figure taken from [33].



Figure 6. Description of a PAT-cell. Figure taken from [33].

The PAT-cells are plugged into a PAT-tester which is placed in a temperature chamber with controlled temperature. The PAT-tester is connected to a PAT-controller which is connected to the computer with the software. By the setup shown in Figure 7, all tests have been performed [34].

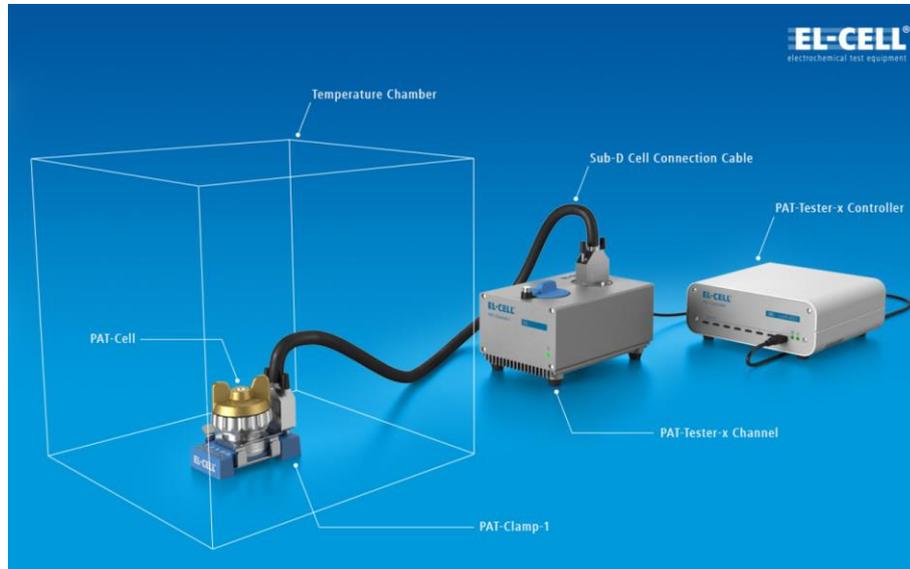


Figure 7. The setup when performing tests with PAT-cells. Figure taken from [34].

2.3 Fast-Charging

Charging of a battery cell is a chemical process that is based on redox reactions. Redox is a combination of oxidation and reduction. The oxidation occurs at the anode and releases electrons which pass through a circuit into the cathode which is then reduced. This process releases energy and is what we define as charging. The number of electrons released this way is measured by the current of a cell in amperes, more electrons released leads to higher current [19].

Fast charging is a concept in which the current is increased to numbers larger than what is normally used. This is normally measured in C-rate where 1C is the current which is needed to fully charge the battery from empty to full in exactly 1 hour. Generally fast-charging covers charging speeds larger than 1C and can go upwards to 6C, which is generally not recommended outside of testing facilities as this can lead to unpredictable reactions inside the battery. Higher C-rates lead to higher temperature development, a battery at 20 °C can at C-rates of 5C or higher reach a surface temperature of upwards 50 °C while C-rates of 1C or lower rarely surpass 30 °C [35].

The availability of fast-charging is something that is generally restricted since the high charge-rates require large amounts of power. An EV battery of around 20-60 kWh that is charged with 2C would require an output of 40-120 kW from the electricity grid which is something that no ordinary household is able to supply [7].

A lot of research has been done by Tomaszewska et.al [36] which summarizes 281 articles, all relating to fast-charging of Li-ion batteries.

Some research includes but is not limited to:

- Packing of cells as well as the ratio between cathode and anode, plays a vital role in battery capacity and has been highly tested [37], [38], [39]. Generally, if the ratio between the negative electrode against the positive electrode is slightly higher than 1 that will favor higher capacity. At higher charging speeds the negative electrode reduces more quickly and therefore increasing the amount can allow for more degradation to take effect without affecting the capacity as much [36].
- CC-CV charging on LiCoO₂/graphite cells done by Zhang et.al [40]. They conclude that at C-rates of 0.4C and above, charging time started to decrease by smaller margins than with C-rates of 0.4C and below. With C-rates of 0.4C or above the risk of lithium plating is increased. The risk to reward ratio is thus lowering with higher C-rates.
- Pulse charging compared to CC-CV charging on LiCoO₂/graphite cells compared by Li et.al [41], concludes that pulse charging can increase both battery lifetime and shorten charging time.
- MCC-CV is tested by Mathieu et.al [42] where current is increased during early stages of charging while slowly decreasing until the battery is fully charged. Mathieu et.al [42] have experimented on MCC-CV where some of their results show an improvement from a standard CC-CV by decreasing charging time while also achieving a longer battery lifetime. They argue that both aspects can be improved by further optimizing the charging protocol.
- Abdel-Monem et.al [43] tested several different protocols on 7 Ah LiFePO₄ such as CC-CV, MCC-CV and more. Results show that more complex charging protocols such as MCC-CV are preferred since they show to retain capacity about 10% better than a CC-CV protocol.

One common conclusion reached by all of the above research is that there is a general lack of testing and knowledge sharing making it hard to claim any results as definitive. Charging of a certain protocol can yield different results depending on the battery chemistry, C-rate, and temperature.

Current derating depending on battery capacity during aging seems to be an untested concept which could lead to improvement in battery lifetime. More complex FCP give results which decrease charging time and increase battery lifetime. A derated current based on battery capacity should lower the stress applied to the battery cell as it ages, slowing the aging process.

2.4 Battery aging

There are two ways to talk about aging in battery cells. One is calendar aging, which consists of all aging processes not included in charging/discharging [44]. The other is cycling aging, the amount charge/discharge cycles the battery cell should be able to withstand [14].

Aging of the battery can be measured by capacity retention which directly shows the current capacity of the battery in relation to its original capacity. This however does not take into account all the different parameters of battery degradation and therefore a better concept of measuring battery cell aging is SOH (state of health). To get accurate measurements of SOH it is important to compile data from usage of the battery cell since single parameters do not always directly correlate to its health [14].

Some of the parameters which cause degradation effects in the battery cell are:

- Thermal decomposition
- Lithium plating
- Mechanical degradation

Performance of the cell goes down for each charging done, every time the battery is used for its intended purpose, when the temperature is too low or too high, from physical wear and tear, and finally, just aging [45].

This experiment mainly treats the effect of fast-charging and the faults arriving from this process which is why focus lies on minimizing those faults as a whole. An analysis, however, is to be made where it is to be noted what degradation effects have taken place.

2.4.1 Thermal decomposition

During cycling of a battery cell there will be thermal generation, either reversible or irreversible where the reversible heat generation don't affect battery lifetime but the irreversible does. At lower C-rates reversible heat generation is more likely, irreversible heat generation is instead linked to higher C-rates and becomes more dominant the larger the C-rate is.

Depending on the cell composition, the rate of where irreversible heat generation becomes more likely varies but, values of 0.5C or below, generally favors reversible heat generation. Battery lifetime is almost twice as long if the average temperature of a battery is lowered from 35 °C to 25°C [46], [36].

Challenges that arise with temperature control of a battery cell are mainly due to the inhomogeneity of the cell pack. The battery cell consists of different materials with different heat conductivity, these materials are not always uniform throughout the cell pack which causes the temperature to develop more quickly in some locations than others. The locations where the current collectors are placed usually have a higher current density which leads to a higher temperature development. Uneven temperature distribution can lead to some spots experiencing localized degradation which can decrease the cells performance overall [36].

Figure 8 show the temperature distribution of a pouch cell where it can be observed that the distribution is uneven throughout the cell. As seen in the figure the temperature is at its highest close to the current collectors.

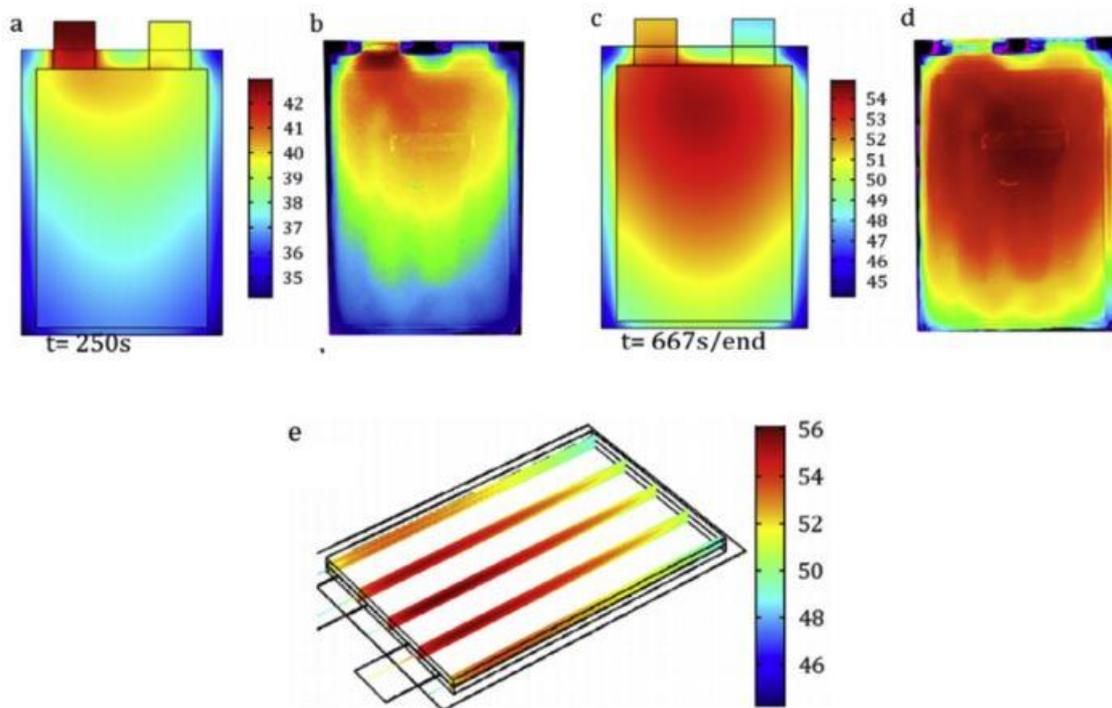


Figure 8. Temperature distribution of a pouch cell. A and C are simulations while B and D are observed from real testing. Figure taken from [36].

The temperature is correlated to many different degradation effects inside the Li-ion batteries. As previously stated, the SEI layer formation is affected by which temperature the formation occurs as well as during cycling. SEI growth during cycling causes the undesirable effect of lithium consumption, meaning binding more lithium to the SEI-layer, lowering the capacity of the cell. Increased thickness of the SEI also leads to a blockage of pores in the layer, impedance will increase at those locations causing a localized temperature elevation which puts the cell integrity in danger [36].

The electrolyte is also sensitive to higher working temperatures. Reactions of the carbon-based solution can create a lot of different products from carbon monoxide to ethylene and alkyl fluorides. PF_5 is a decomposed product of the LiPF_6 -electrolyte and occurs at high temperature. The PF_5 has a dissolving effect on the SEI which exposes the graphite anode surface to other decomposed products. The decomposition of LiPF_6 is triggered above $50\text{ }^\circ\text{C}$ and is more prevalent the higher the temperatures. This rarely happens and is most prevalent under thermal runaway [36].

Thermal runaway is probably the most disastrous result that can be achieved by looking at temperature decomposition. Not only does it entail major safety hazards but also renders the cell completely unusable after its occurrence. The cause of thermal runaway is when the SEI-layer is damaged by deterioration (physical or chemical) and exposes the anode to decomposed electrolyte products. This produces an exothermic reaction to take place, leading to an increased temperature development, triggering more reactions throughout the cell which eventually short-circuits the cell.

The electrolyte will decompose during this massive temperature of around $250\text{-}350\text{ }^\circ\text{C}$ causing hazardous vapors to build up inside the cell module, increasing the pressure and finally leaking out [36].

Solutions to high temperature exist by utilizing different cooling techniques. Liquid cooling is seen as the more useful tool and can be upwards of 3500 times more efficient at cooling battery cells compared to air cooling [48]. Experiments have shown that battery cell temperature can be controlled under 40 °C at a C-rate of 5 depending on the material and technology used [47].

2.4.2 Lithium plating

Lithium plating is another very common reason for battery degradation. This phenomenon is more likely to occur at low temperatures, high charging rate, non-uniform charging or if the anode potential dips below 0 V. Anode potential can be hard to observe in a commercial cell since potential is usually measured in dynamic voltage and finding the potential of each separate electrode is not possible.

The process of lithium plating is a formation on the anode material where the Li^+ which are supposed to move from the cathode through the electrolyte and then intercalated into the anode, instead causes a buildup of lithium on the anode surface. The lithium buildup then reacts with electrolyte which consumes both active lithium and electrolyte, leading to not only battery degradation but also an increased anode polarization which allows for favorable lithium plating conditions [48].

2.4.3 Mechanical

Mechanical degradation is any form of physical damage done to the cell. Examples of physical damage include cracks in the electrodes, loss of connection between current collector and the active material and similar. There are no reactions involved but rather effect caused by stress in the battery caused by current.

During charging, the Li-ions should intercalate into, and exit, the graphite. To keep the battery healthy this process should be uniform but at high C-rates the Li-ion concentration gradient can vary a lot, causing the anode material to be unable cope with the situation. This can lead to cracking in the electrode and further on, cracks in the SEI. Cracking leads to the exposure of the anode active material, leaving it to react with fresh electrolyte and forming more SEI which is going to be uneven, increasing the risk for further problems such as poor conductivity or uneven temperature development [36].

2.5 Fast-charge protocol

A FCP or fast-charge strategy is a way to describe a charging process. A charging process entails everything that is done to the cell during charging, from adjusting the current levels to regulating the temperature of the cell. Most common and simplest is the CC-CV protocol which charges with constant current (CC) until it reaches the desired voltage in which it keeps the voltage constant (CV) until the current has dropped to the desired cut-off level.

A FCP can be created in many different ways with several parameters in mind. It could be temperature dependent where it tries to relate the current based on the surface temperature of the cell. It could utilize pulse-charging in which you charge with maximum current for a short period of time and then shut it off for a short period of time, repeating this pattern until the battery is charged to max [36].

In many cases the FCP is presented in terms of state of charge levels (SOC). SOC is a measurement which shows the amount of electrical energy stored in the cell. This is related to

the dynamic voltage level of the battery, but it's not directly related. Voltage measurements are needed and getting the exact correlation takes a lot of resources which makes SOC an efficient way for cell producers to present information of their battery without exposing their data.

2.6 UN Sustainable Development Goals

2.6.1 SDG 9: Industry, innovation, and infrastructure

The increase of electric cars on the roads also comes with infrastructural changes. The amount of charging stations needs to increase and develop in both urban, sub-urban and rural places for the integration of EVs to be successful. Many different options on how to charge the cars in the most beneficial way have been investigated, such as charging through the road, with overhead-lines or charging stations at the parking lot where the one mentioned last is the most common one [49].

Today's infrastructure is built for ICE-vehicles thus a majority of the society have the same expectations for EVs when it comes to driving range and the ability to drive long distances [50]. Highways in the current infrastructure often stretch over very long distances, for an EV to travel over these distances a bit more planning can be necessary. For example, check where there are charging stations and then plan when to stop and charge the car. A study has been made where EVs are driven at highway speeds in Australia in different scenarios. For one of the cars tested, the manufacturer had published a drivable range of up to 199 km, however the study showed that the EV could only be driven up to 70 km when driving at the speed of 110 km/hour. There are a lot of factors affecting the range of an EV such as battery degradation, charging strategies and driving patterns. For example, by planning when to fast-charge, long distance travel could be easily feasible [50]. It should be mentioned that depending on the everyday travel needed, majority of drivers can perform their everyday driving with an EV without bigger adaption [15].

2.6.2 SDG 11: Sustainable cities and communities

Making our cities and communities more sustainable is a vital part of reaching a more sustainable future. Within this category, both buildings, infrastructure and transportation are included and need to be considered. Already now, 70% of global carbon emissions are produced by cities [6] and United Nations Development Program, (UNDP) expect that two thirds of the population will live in urban cities by year 2050, which is about 6.5 billion people [51].

A big part of this is the transition to a more sustainable way of transportation. In today's society, a majority of cars are still conventional ICE-vehicles fueled by fossil fuels. By reducing the use of the conventional cars, the emissions from burning fossil fuels will decrease. When burning fossil fuels, different elements and gases are emitted into the atmosphere which could lead to environmental damage. The main component of combustion is CO₂. Other components include Sulphur oxide (SO_x) and nitrogen oxide (NO_x) [1].

There are a few different ways to reduce the use of the conventional ICE-vehicle, one way is to replace it with some other sort of transportation. This could be done by riding a bike, using public transportation more often or exchange the car into an EV. One of the main options in the public transportation are buses. These can be both electricity-driven as well as run by fossil fuels. In the latter case, it is still a better option than if all the passengers would have taken a separate car to their destination. In some urban cities in European countries, many of the buses

are electrically driven (also called EBs) and the use of EVs have increased a lot during the past few years [52].

For this change in transportation to be possible, it is very important to develop sustainable, reliable, and durable batteries which can withstand many cycles of charging and discharging without being too degraded. If batteries start to degrade too soon and need to be replaced often, they will turn out to be a less sustainable choice due to the material use and waste management [51].

2.6.3 SDG 13: Climate action

Sustainable development goal number 13 refers to meeting the climate targets that have been set during the past years to prevent further damage of the environment as a result of the industrialization of societies. The emissions of greenhouse gases have increased by a lot during the past decades and have already led to multiple sorts of damages on the environment, for example the ozone layer. The main component in the emissions of greenhouse gases is carbon dioxide, which is a result of burning coal, oil, and other fossil-based resources. The transition to the electric car could therefore have a positive impact for this goal as well, at least in a country like Sweden where most of the energy system is supplied by renewable resources [1]. However, in most parts of the world, majority of the electricity produced is from fossil-based resources, and in those cases the transition to EVs might not have only a positive impact on the environment still.

3. Method

The method used during this thesis project is focused on an experimental investigation. Different cycling tests have been performed on two different battery cell formats, pouch cells and PAT-cells. The first part of the testing was performed on four pouch cells, based on Li-NMC/graphite chemistries, which are provided by Volvo Cars. These cells are referred to as the commercial cells later in this report. The tests have been performed at Volvo Cars Battery Lab. Our main contribution has been to write the test protocols and fast charging protocols (FCP), together with two other students from another thesis group. A FCP describes the process of charging the cell from a SOC level of 0% to a SOC level of 100% and includes charging rates (C-rates) variation, as a step function, to assure that cell operations are kept within the specified operational voltage range.

The test protocol is written following Volvo Cars standard protocol, in which specific boundaries, for example voltage levels and temperatures, are set. During testing, the cycles record the voltage and current of the cell versus time. A more detailed explanation of the FCPs is presented in section 3.1. This was also done together with two other students.

The second part of our thesis project is to perform the same tests on smaller scale in which PAT-cells from the company EL-cell are used. EL-cell testing can be done in several different ways due to their software allowing for different cell testing conditions. In this project, three-electrode cell (PAT-cell) testing has been conducted to measure cell and electrodes voltage simultaneously. This is not possible with the commercial cells. The EL-cells testing is divided into several different scripts. Each script consists of steps which describe one part of the cycling. The steps can have different cycling conditions, for example charging with a constant current until the cell voltage reaches a cut-off value, or it can also be discharging at certain rate until enough time has passed. These steps complete a script, and several scripts are being executed to best copy the same testing done on the commercial cells.

The same FCPs have been used on both the commercial cells and the EL-cells. Due to project time limitations, the two cells' formats show a difference in the number of completed cycles, where the commercial cells have been cycled for 350 cycles whereas the EL-cells have been cycled for 100 cycles.

The tests that have been performed on both the commercial cells as well as the EL-cells will be explained in section 3.1 and 3.3 respectively. Further, a tear down of a cell of the same sort as our commercial cells, have been performed where the cell is disassembled and will be described in section 3.2. The material extracted from the electrodes of the disassembled cell has been used to build the PAT-cells, and the construction of these is explained more in detail under section 3.3.

3.1 Testing of commercial cells

When starting the testing of the commercial cells, 50 cycles were setup as a trial test to validate the protocol script. During these 50 cycles however, the FCP was not followed for all the cells, and the cycling did not go as planned. After these 50 cycles, the capacity of the battery cells had gone down by only 0.5-1.3% for all four cells. Due to the difference with the FCP later applied, the 50 cycles are not considered as a part of the results and are not counted for when mentioning on the total amount of cycles performed.

Since four cells are available for testing, it was decided to focus on two different FCPs, thus we had two cells for each FCP which allowed for better accuracy of the results. For all commercial cells, the battery cells are cycled for 350 cycles (excluding the 50 test cycles).

In between every 50 cycles, a fast reference performance test (RPT) is performed. The RPT is performed as a periodic assessment of battery degradation during cycling. Through the RPT it is possible to measure the capacity fade, and resistance rise as a function of test time. The RPT is the same for all four cells and consisted of four different steps. The first is a formation step which is done to activate the cell properly through a charge-discharge cycle. Next step is a capacity step where the actual capacity of the cell is measured. The third step is a pulse test which is done to measure power and resistance fade through pulses at different SOC levels and the last step is to adjust the SOC-level that is valid for transportation and storage.

Table 2 is an example of a FCP where the SOC-level (%) is related to the C-rates and cell power (W), which is the current multiplied by the cell voltage. The exact values of SOC-levels and C-rates are specified by the supplier are not presented since that is a proprietary of Volvo Cars.

Table 2. An example of a FCP where the SOC is related to the cell power. Disclaimer: Specified SOC-levels and C-rates by the supplier are a proprietary of Volvo Cars.

SOC (%)	0	100
C-rate (stepfunction)	
Power (W)	321	386	378	364	349	335	319	304	288	272	256	241	225	209	173	141	

In an FCP, a current value is specified for each predetermined SOC value for one cycle. The strategy to associate current to its specific SOC value is a proprietary of Volvo Cars. In the first FCP, referred to as FCP 1, the value of the current is specified for each SOC value and does not change in between cycles. Hence, all 350 cycles are run with the same current values for each SOC level. This is the standard FCP used by the supplier of the battery cells and was used as a reference case. The second FCP, referred to as FCP 2, has a constant C-rate but the current is adjusted according to the capacity after every 50 cycles. This is done by using the capacity from the capacity check at each RPT. After 350 cycles, the tests are stopped and later a tear down of cell 2 and cell 3 respectively, is performed. The teardown method is presented and discussed in the following section, 3.2.

3.2 Teardown of one commercial cell

To be able to construct the EL-cells, a teardown of a pristine cell has been performed, this cell is of the same type used in previously mentioned testing of commercial cells. The teardown has been done together with a lab engineer to get the material for the anode and cathode used in the EL-cells.

During the teardown, the cell is opened in a glovebox which is a big, sealed container filled with argon-gas. The levels of oxygen and water need to be as low as possible to prevent any deterioration of the material in the cell, due to possible reactions with the electrolyte and lithium. For the day of the teardown, the oxygen level was varying between 0-0.8 ppm and the moisture level was varying between 0-13.7 ppm which is acceptable but still a bit high. Moisture being captured inside the cells can cause problems such as short-circuiting depending on the amount. Oxygen reacts with electrolyte causing it to degrade even before being cycled.

The cell used is a pouch cell. The electrodes and separator contained in the cell have the shape of long sheets and are stacked in many layers. In every sheet, there is a current collector which is coated with active material, which is either the anode or the cathode. The active material needs to be scrubbed off, to expose the current collector, for this ethanol of 96% concentration is used. The current collector on the anode is copper, while the one on the cathode is aluminum. When a big enough surface is cleaned, approximately 7x7 cm, electrodes could be cut out. As a reference, these electrodes are in circular form with a diameter of 18 mm.

Furthermore, two of the cycled cells are also open. One from FCP 1 (cell 2) and one from FCP 2 (cell 3). This is to try and observe any degradation effects on the battery and compare them between each other. We study the surface of the anode by looking for any spots which can indicate lithium plating, this is very hard to do since lithium plating is hard to detect and can come in small sections. The two cycled cell are then measured in a scanning electron microscope (SEM).

3.3 Construction of PAT-cells

The construction of PAT-cells is performed in a glovebox. To understand and get used to building PAT-cells, two dummy cells are constructed. These cells are also used to try out the EL-software to make sure that it is correctly setup to perform the real testing and that the charging profile would follow our protocols correctly. After the test on dummy cells had been completed, the real PAT-cells are put together with the material obtained from the tear down. The assembly has been made by following the given manual from EL-cell. An illustrative image of the assembly process can be seen in Figure 9.

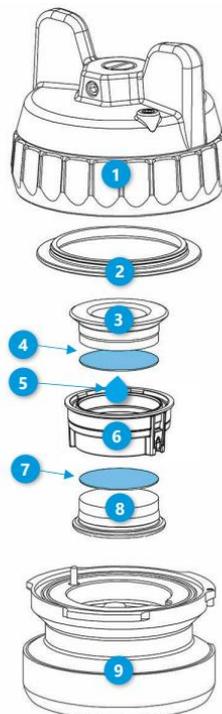


Figure 9. Illustrative image of the assembly process of the PAT-cell. No.1: Screw cap, no.2: Sealing ring, no.3: Upper plunger, no.4: Upper electrode, no.5: Electrolyte, no.6: Insulation sleeve, no.7: Lower electrode, no.8: Lower plunger, no.9: Cell base. Picture taken from [33].

When constructing the PAT-cells, the manual is followed closely as every step is explained in detail. Starting with the bottom half cell, the cathode electrode is placed in the PAT-core with the active material facing the reference electrode in the insulation sleeve (part no. 6 in Figure 9). The bottom is then closed and sealed with a lower plunger and placed in the cell base. On the upper side of the reference electrode, in the insulation sleeve, the first layer is soaked with electrolyte. In these PAT-cells, 100 μl of electrolyte is used. On top of the soaked area, the anode electrode is placed with the active material facing the soaked surface and the upper plunger is attached. The two half cells are then mounted and sealed with a lid, creating one full cell. Four full cells were built using the same material and the same amount of electrolyte. They are built during the same day in which the level of oxygen was 35 ppm and water was 8.7 ppm respectively.

3.4 SEM & BSE

After the teardown a scanning electron microscope (SEM) is used to verify any degradation effects on the anode. The SEM sends electrons out in a beam against the material where the electrons interact with its atoms, returning a signal which is then registered by the SEM. This signal is measured and used to produce an image of the surface of the anode [53].

This is a very useful tool to detect any growth on the anode which should mainly be lithium plating, the SEI could have some growth as well but should not be as extreme as the lithium plating. We can use this together with the backscatter electron (BSE) technique which measure electrons returning from another angle. These electrons penetrate further into the material and give a measurement of what atoms appear in the scanning [54].

4. Results and Discussion

In this section the results for all four commercial cells are presented and discussed. Results from the four cells are displayed for comparison in all figures, they are named cell 1, cell 2, cell 3, and cell 4 and follow a consistent color-coding. The results have been evaluated using a software called Concerto AVL together with Python, while being plotted in MATLAB.

In section 4.5 all results and parameters are summarized in Table 3, for an easier and clearer comparison. Further, the effects of degradation are discussed and evaluated by presenting figures from the SEM-analysis.

4.1 Commercial cells – Capacity retention

The cells tested with FCP1, cell 1 and cell 2, are for every cycle both charged in several steps with C-rates that decreases as a function of the SOC of the cell. The C-rates are determined by supplier, set at the start of testing, and do not change between cycles. The cells for FCP2, cell 3 and cell 4, are also charged with C-rates that decreases as a function of the SOC of the cell. The C-rates are kept constant throughout the testing, but the capacity is updated each 50 cycles during the RPT, leading to the current adjusting accordingly.

In Figure 10, the capacity retention is plotted against the number of cycles for all four cells. The dashed lines that are red and pink, are fast charged according to FCP 1 while the two blue graphs are fast charged according to FCP 2. The capacity has been measured after every 50 cycles during the RPT.

Figure 10 shows clearly that there is a higher capacity loss, indicating a higher degradation, for cell 1 and 2 than for cell 3 and 4, since cell 3 and cell 4 follows FCP 2 with adjusted currents according to the actual capacity. By adaptively reducing the current flowing in the cells, the damage on the battery is decreased.

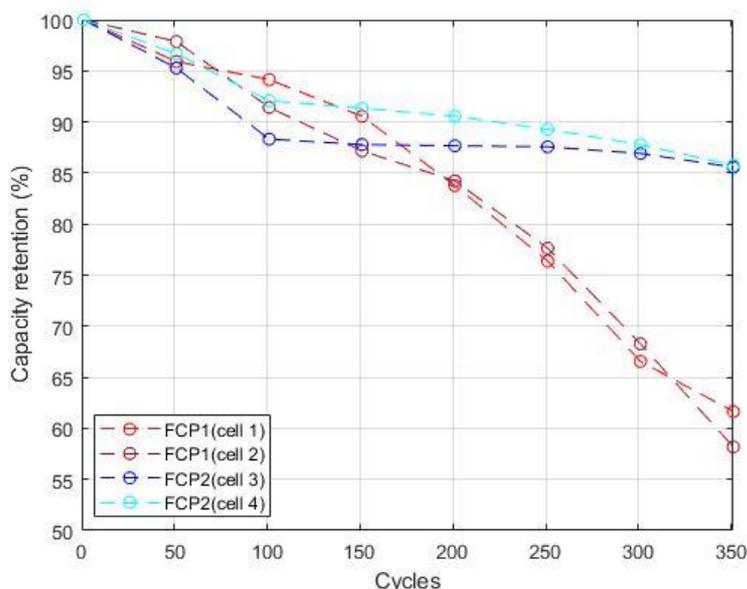


Figure 10. Capacity retention (%) related to the number of cycles for every 50th cycle from 1 to 350, for all four cells.

For FCP 1 it is evident that both cells have experienced a significant degradation and would have reached their end of life (EOL) around 275 cycles (considering an EOL = 70-80% capacity

retention). The results show that the two FCPs have similar effects on the cells until 90% capacity retention. The capacity degradation is gradually increasing in FCP 1 the more cycles have been completed. This indicates that keeping the fast-charging protocol constant, without adapting the current to the variation of the capacity will accelerate aging of the battery.

The capacity retention of FCP 2 shows significant improvement after 150 cycles with respect to FCP 1, and the loss of capacity for FCP 2 after 350 cycles is only around 14.5%. The curve is flattening as cycling is progressing and between cycle 100 to 350, we only see about a 3-4% decrease while the decaying from cycle 1 through 100 was around 12%.

The results confirmed the hypothesis of fast charging according to FCP 2, as it clearly shows positive effects in reducing the aging of the battery. The high decrease in capacity for cell 1 and cell 2 respectively, could be due to several reasons, such as a progressive damaging of the electrode materials, parasitic reactions limiting the available lithium left in the electrolyte and build-up of lithium plating. Detailed investigation of the degradation phenomena is out of the scope of this thesis but will be further discussed in section 6 Future Work.

4.2 Commercial cells – Energy throughput

The energy throughput is the amount of energy flowing in the battery when the battery is charging or discharging during one cycle. The energy throughput has been evaluated with regards to charging, and Figure 11 presents how the energy throughput while charging is changing during the cycling for all four cells. The charging rate is in watt-hours (Wh) and has been calculated for every 50th cycle from cycle 1 to 350.

Since the capacity retention has clearly decreased more for cell 1 and cell 2 than for cell 3 and cell 4, the results of the energy throughput should follow the same trend. By that, the cells fast-charged according to FCP 1 should have a lower energy throughput at the end of cycling, than the cells fast-charged according to FCP 2.

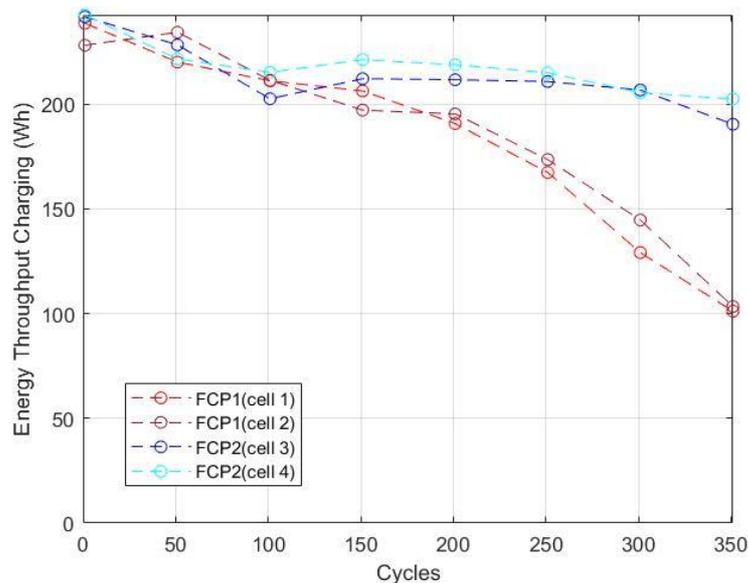


Figure 11. Energy throughput (Wh) when charging for every 50th cycle from 1 to 350, for all four cells.

As seen in Figure 11, the amount of energy throughput while charging is decreasing quite rapidly for the cells charged according to FCP 1 (cell 1 and cell 2), especially after 200 cycles.

This could be taken as a sign of a decrease in the capacity of the battery due to degradation, which is proven and shown in the previous Figure 10. Cell 1 shows a bit lower energy throughput in the last 100 cycles than cell 2, however they both are decreasing by more than half of the initial capacity.

However, for cell 3 and cell 4, the amount of energy throughput while charging is not decreasing much. The amount of energy throughput while charging is decreasing a bit during the first 150 cycles but not as much in the remaining 200 cycles. This strengthens the capacity retention result, that the two FCPs have similar degradation effects for the first 100-150 cycles.

The last 200 cycles are relatively steady for cell 3 and cell 4, which indicates that the capacity of the battery cell is still relatively high at the second half of the cycling (which is the case as seen in Figure 10). Cell 3 shows a higher decrease in the beginning but an increase in energy throughput after 125 cycles and keeps it relatively steady until the end. Since values have been taken for specific, individual cycles, some values could be a bit off. Therefore, it is the trends that are most important. Cell 4 has a bit more variation with the amount of energy throughput for certain cycles, however, still has a higher amount flowing through the battery cell in the end.

4.3 Commercial cells – Charging time

In Figure 12, the time (s) it takes to charge the cell during a cycle is presented. The charging time have been taken for every 50th cycle and is evaluated for all four cells. The expected outcome is that the individual cells follow the same trend as in capacity retention and energy throughput. It is therefore expected that cell 3 and cell 4 will have a higher charging time by the end of the cycling, than cell 1 and cell 2.

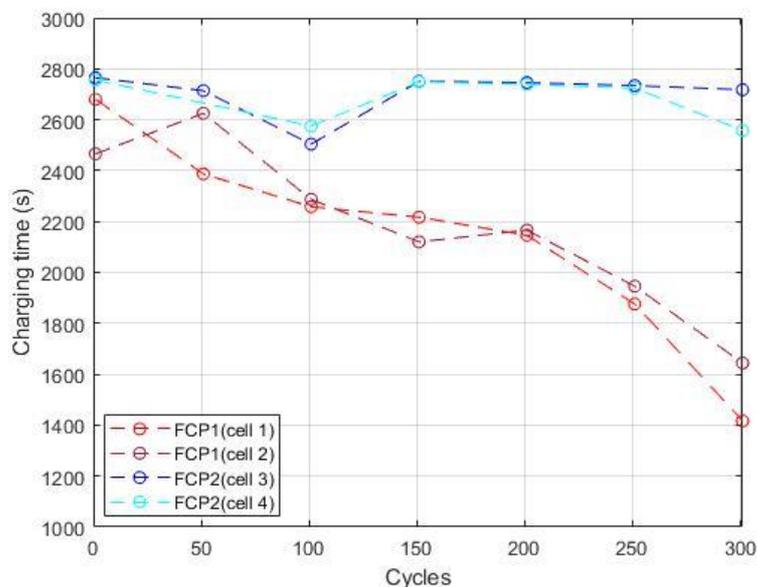


Figure 12. Charging time for the cells to fully charge, taken every 50th cycle from cycle 1 to 350, for all four cells.

As expected, charging time is following a similar trend to the capacity retention and energy throughput. A decrease in charging time means that the battery is taking shorter time to charge to max. The percentage drop-in charging time of cell 1 match well with the capacity retention

where cell 1 lost around 30-35% capacity while charging time decreased by the same amount. This follows naturally as the FCP 1 doesn't consider any aging effect during cycling.

Cell 2 however shows a little deviation from the trend in capacity retention, where the capacity loss was similar to cell 1 but charging time decreased around 45% instead. This could be attributed to the measuring equipment which is used. Since the current is based on SOC levels and these levels could be separated by small margins it is plausible that the voltage could shift a bit during decreased current step.

For cell 3 and cell 4 the charging time decreases from 2800 seconds to about 2500-2600 seconds in the first 100 cycles. This is easily explained by the large decrease of capacity during this period and the fact that capacity is only updated each 50 cycles. During that interval the degradation is progressing with such rate that the charging time have noticeable variations in between each cycle. A quick analysis has been made which show that charging time between cycle 1 and 25 decreased by 150 seconds and 100 seconds between cycle 51 and 75 for cell 3 and cell 4 respectively. After the large degradation in the first 100 cycles the curve starts to straighten out which is in line with the expected results. Since C-rate should be constantly related to an updated capacity and when the capacity variations between each 50 cycles are small, so is the charging time variations. When the degradation is large, variations in cycling time is to be expected, when the degradation is lower, variations in cycling time is also lowered.

4.4 Commercial cells - Resistance

During the RPT between the cycling, pulse tests are done to measure the resistance and impedance of the cells. The method provides a direct-current internal resistance (DCIR) value which is defined as the sum of pure ohmic resistance and polarization resistance. It can also be used to get a good indication of the available power of the battery and power losses.

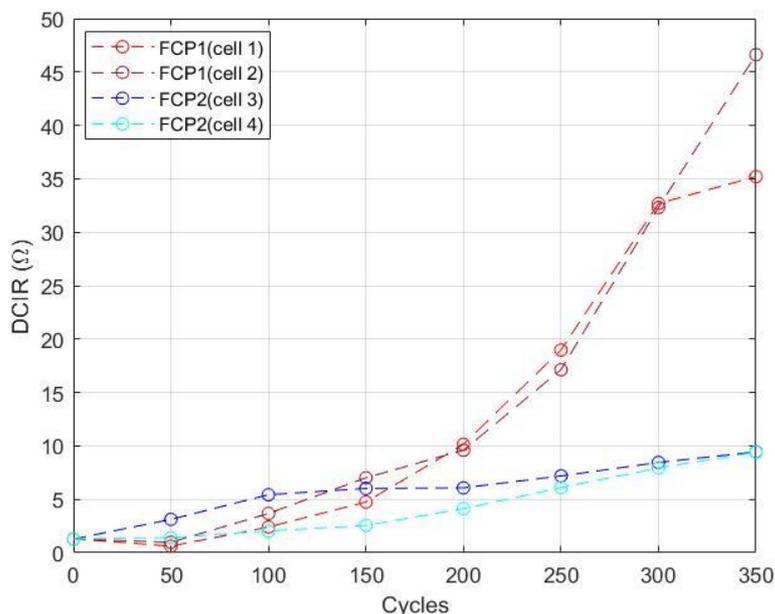


Figure 13. DCR for FCP 1 and FCP 2, measurements are evaluated for one pulse during the RPT.

All four cells started off similarly but at 150 cycles they began to deviate from one another with FCP1 seeing a larger increase of the DCIR compared to FCP 2. This is again another confirmation of the similar effects of the two FCPs in the first 100-150 cycles, and subsequent

stabilization of performance of the cells tested according to FCP 2. The electrochemical reasons behind this behavior should be further investigated.

As can be seen from Figure 13, the DCIR is increasing for all cells and indicates a decrease of power capability meaning that the battery is unable to deliver energy at the same rate after 300 cycles compared to the start. Ohmic resistance is heavily dominated by electrolyte in battery cells [55] and since the DCIR method is built on ohmic resistance, an increase in DCIR seems to indicate that the electrolyte has been degraded.

4.5 Summary of the parameters and results of FCP 1 and FCP 2

In this section, the results are summarized and discussed with regards to the results from the second tear down after the cycling, and the SEM-analysis which is presented in section 4.6. The results from the tear down are presented in figures as well as the results from the SEM-analysis. These are expected to show increased degradation of cell 1 and cell 2, compared to cell 3 and cell 4. In Table 3, the capacity, energy throughput, charging time and DCIR are measured at the first as well as the last cycle, cycle 1 and cycle 350 respectively. These are evaluated and compared with each other. Capacity retention, energy throughput and charging time show a decrease from cycle 1 compared to cycle 350. DCIR show an increase in the same interval.

Table 3. Summary of the capacity retention, energy throughput, charging time and DCIR in percentage where the measurement of the last cycle is compared to the first cycle for cell 1, cell 2, cell 3 and cell 4 respectively.

<i>Parameter/Cell-ID</i>	<i>Cell 1</i>	<i>Cell 2</i>	<i>Cell 3</i>	<i>Cell 4</i>
<i>Capacity retention</i>	61.65%	58.19%	85.57%	85.76%
<i>Energy throughput</i>	42.38%	45.33%	78.77%	83.43%
<i>Charging time</i>	52.87%	66.67%	98.3%	92.82%
<i>DCIR</i>	2712.97%	3659.14%	736.91%	732.95%

First thing to note is that there are variations between the cells cycled with the same FCP. Cell 1 and 2 have a slight change in capacity retention and energy throughput while charging time and DCIR measurements show large differences.

Charging time variations can be explained by the programming of the charging steps and cell behavior. The programming is set to change current when reaching a specific voltage during cycling, when the current is lowered the stress on the cell is lessened which can cause the cell voltage to spike [56]. The system can detect that change and if the voltage is high enough, it can then change into the next charging step essentially skipping one step. These events are not always triggered at the same times but rather occur randomly and can vary charging time significantly.

DCIR is measured during the RPT with pulses and can also vary but only slightly. The largest variations between the two cells are happening in the last 50 cycles of the test. It is hard to give any accurate explanations since there is only one occurrence of this. One explanation is that cell 2 shows signs of polarization at that time, causing a voltage drop and a decrease in resistance in that point [57].

Cells 3 and 4 also show slight variations but mostly in energy throughput. This can be explained with the same reasoning as above regarding charging time. Cell voltage varies as current changes, and this affects not only charging time but also energy throughput.

4.6 Results from tear down and SEM-analysis

During the second tear down when cell 2 and cell 3 are disassembled, the sheets within the pouch cell are investigated to see if there are any degradation effects strengthening the previously mentioned results. What is expected to see is more stains and spots on the anode of cell 2 comparing with cell 3 since it has higher degradation. Figure 14 shows the anode of a pristine cell. The anode of cell 2, which has been run with FCP 1 is presented in Figure 15. The anode of cell 3 which has been run with FCP 2 is presented in Figure 16.



Figure 14. Anode of a pristine cell from 1st tear down.



Figure 15. Anode of cell 2 after 350 cycles, from second tear down.



Figure 16. Anode of cell 3 after 350 cycles, from second tear down.

Studying the images closely, cell 2 has degraded more than cell 3. In the pouches, cell 2 is very swollen which indicates that the electrolyte reacted chemically causing various gases to build up inside. This is confirmed when the opening of the cells shows only trace amounts of electrolyte on cell 2. Cell 3, although containing more, also shows a significant decay of the electrolyte. To confirm what is suspected, a SEM-analysis is performed which gives better indication of what may be the degrading effect. The SEM is performed in 20 kV and the quantification is in atomic%. Since the SEM gives information about which materials exist in the investigated area based on atomic weights, it gives indications about the degradation effects. If the level of sulphur, fluor, phosphorous and/or aluminum is high, this often indicate that the electrolyte has decayed due to the electrolyte containing these elements. However, the Sem cannot detect which component the elements come from, and therefore only gives indications.

In Figure 17, a zoomed in part of the anode of cell 2 is displayed. The areas marked show which spots have been investigated. The information received about the elements from the SEM are presented in Table 4. It is the levels that give an indication of what could have happened.

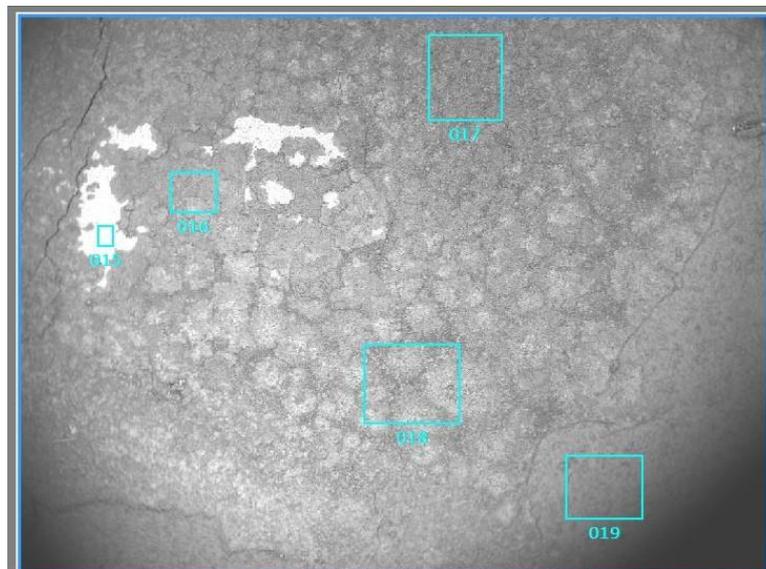


Figure 17. Anode from cell 2, from the SEM analysis. Performed after 350 cycles.

Table 4. Information received from the SEM-analysis when analyzing the area displayed in Figure 17.

Area/Element	C	O	F	Al	P	S
015	22.6	33.8	4.5	37.4	1.8	
016	73.1	15.3	7.3	0.4	3.5	0.5
017	50.4	32.9	10.4	1.3	4.4	0.6
018	50.6	32.2	10.6	1.4	4.6	0.6
019	69.4	20.8	6.5		3.3	

When there is a clear sign of lithium plating, there is a clear structure, which is not seen in neither of the cells. However, the SEM analysis does indicate some growths on the anode for cell 2 but not as large growths for cell 3. The BSE confirms large amounts of aluminum oxide over both anodes as well as residue from solvents used in the electrolyte. Some of the small growths confirmed low amounts of aluminum but large amounts of oxide which is usually a

sign of lithium plating. Lithium cannot be detected by the BES due to its low atomic density which is why large quantities of oxide without any other element is an indicator of lithium plating. The SEM did show that the main degradation effect is the decay of electrolyte as can be seen in Table 4. Most of the areas investigated showed the same results, and therefore only one has been chosen to be displayed.

4.7 Discussion about the results of the commercial cells

The results from the testing of the commercial cells, show significant differences when fast charging according to FCP 1 versus FCP 2. The main parameters investigated are the capacity retention, energy throughput, charging time and DCIR (direct-current internal resistance) and these are summarized in Table 3. Since all parameters have shown similar effects, it seems to be a great degradation effect when fast charging according to FCP 1. The degrading effects in FCP 1 are shown for both cell 1 and cell 2, this indicates that the results obtained are due to the fast-charging protocols and not because of cell variations.

The results of the cells fast charged according to FCP 2, cell 3 and cell 4, have shown much slower aging of the battery and higher capacity retention, than cell 1 and cell 2, as is expected. This indicates that adjusting the current according to the varying cell capacity, have positive effects on the durability of the battery. When performing the tear down, the results are strengthened as the cells disassembled show high differences in degradation. Cell 2, fast charged according to FCP 1, shows indications of lithium plating, buildup of aluminum oxide and loss of electrolyte. The same effects, but in much smaller scale, are seen in cell 3, fast-charged according to FCP 2. However, the results given are based on certain limitations.

The temperature is kept constant at 35 °C in a temperature chamber to ensure minimal variation. By adjusting the temperature, one can achieve results which differ from the results in this thesis. The temperature that the cells have been cycled in is a bit higher than room temperature to speed up the aging process. In different ambient temperature, the charging process will vary, and therefore this might be of interest. The number of cells tested is also a limitation, the fast-charging protocols have been tested on 2 cells each, which may not be enough to give fully reliable results even if the results do show the same effects for both pairs of cells. By testing on more cells, and different types of cells (not only pouch cells), more reliable and recurring results can be achieved. Another limitation, for FCP 2, is that the capacity has always been calculated for every 50th cycle, by doing the capacity calculations more often or seldom, other, or more evident effects may be seen.

Further, the main factor that limited this thesis project is the time constraint. The battery cells have only been cycled for about 3 months. Even though this experiment has given clear indications after a short period of time, to get more reliable results the tests need to be performed over a longer period.

The results received from this thesis work should mainly be used for further studies within fast-charging strategies for EVs. The results show clear benefits in favor of FCP 2 and by performing further testing on battery cells according to this fast-charging protocol, and minimizing the limitations of the testing, these results can be further strengthened. Implementing FCP 2 in an EV today can be very tricky due to the battery in the car needing to adjust the current and perform capacity checks after a fixed number of cycles. However, if a derating function is extracted after a longer period of testing, an average derating constant of the capacity can be calculated. By then lowering the capacity with this constant, the current can be adjusted accordingly, which might be an easier implementation.

4.8 PAT-cells – Capacity retention

The research on PAT-cells is made to investigate if there exist a possibility that results obtained from large commercial cells can be recreated by using smaller cells with the same material. This will prove to be very useful since more testing can be done by using smaller cells and costs will go down, not only that, but this can also strengthen or weaken the previous results depending on the outcome.

The same FCP's are tried on the PAT-cells as well as RPT 25 °C and 35 °C but with adjusted current values. SOC levels are decided from the first cycle of the commercial cells where an average is drawn from FCP1 and the same for FCP2. The SOC levels are observed to not always increase and thus during dips an interpolation is done to correct those.

Capacity retention of the PAT-cells against cycles are shown in Figure 18. This is done in the same way as previous Figure 10. Only 100 cycles are completed. The capacity is increasing slightly during the first 50 cycles which can be explained due to polarization of the cell and settling of electrolyte inside the electrodes. At 100 cycles the degradation is between 1-7 %.

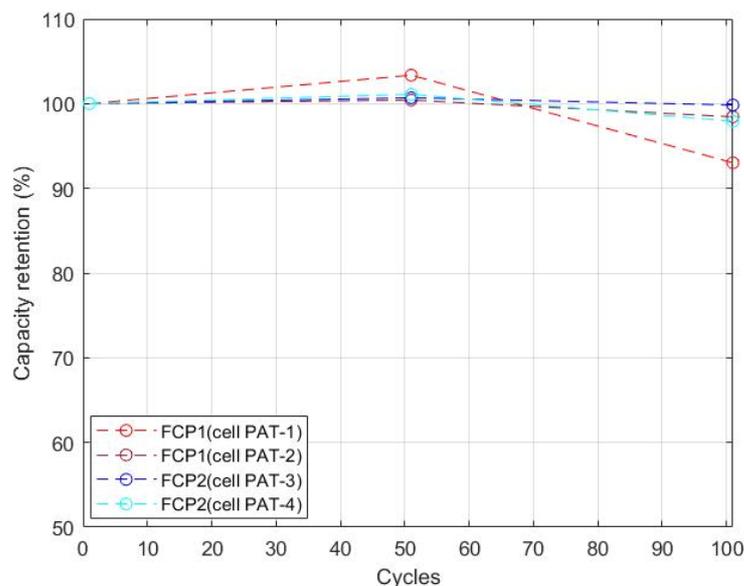


Figure 18. Capacity retention (%) related to the number of cycles for every 50th cycle from 1 to 100, for all four cells.

4.9 PAT-cells – Potential profile

In Figure 19 the total potential, as well as the potential of the anode and cathode is measured during a full charge discharge cycle. A 30-minute rest time is included, both after the charging and also after the discharging. These measurements look very similar to previous measurements as can be seen in Figure 4.

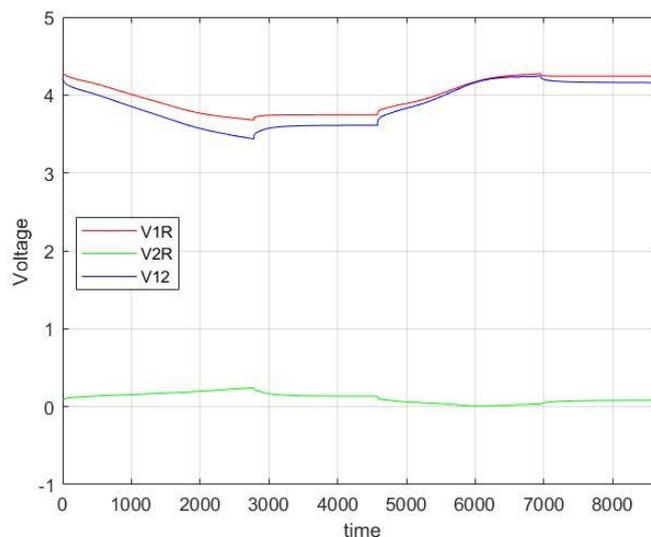


Figure 19. Cell potential of PAT-1 during discharging, rest, and charging. V_{1R} is the potential of the cathode against the reference electrode, V_{2R} is the potential of the anode against the reference electrode and V_{12} is the difference in potential between the anode and the cathode.

Figure 20 show the difference in potential of the anode between the first cycle and the 101st cycle for PAT-1. Here the two cycles look very similar, note that the 101st cycle has been shifted in time to match the first cycle. Zooming in on the charging step of the cycle, the potential can be seen to slightly deviate from one another. During the 101st cycle the anode potential manages to reach negative voltage numbers, indicating that the potential of the cells slightly changes during aging. The same effects can be seen throughout all cells.

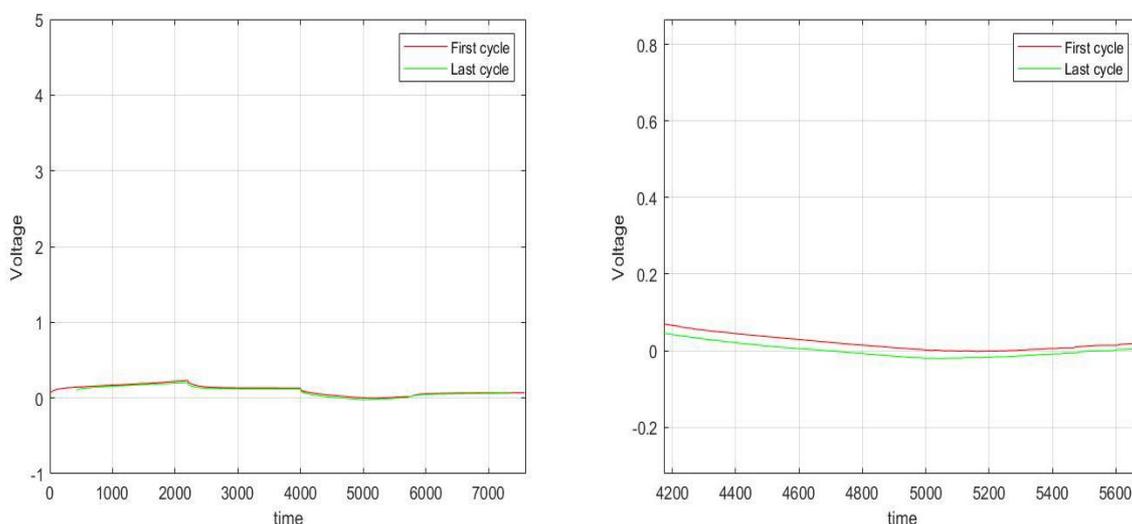


Figure 20. Anode potential of PAT-1 during discharging, rest and charging for the first cycle and the 101st cycle.

4.10 PAT-cells – Discussion of the method, limitations, and results

The results from EL-testing are hard to evaluate due to the few datapoints available. Cell PAT-1 is varying more than the other cells. This can most likely be explained by the many uncertainty factors during the testing. Factors such as the levels of O_2 and H_2O varying close to the limit of the recommended ranges (preferably as close to 0 ppm as possible). This,

together with faulty cycling/testing equipment which interrupts cycling at random points. Many attempts have been done but unfortunately none worked fully after the first 100 cycles. These uncertainties are hard to avoid due to the time restriction given at the start and the tests would most likely not proceed if given unlimited time.

What can be noted from the results is that the potential is acting accordingly to the commercial cells and literature as recalled in Figure 4. This at least indicates that the cells are constructed right and should be able to perform if testing is continued. During aging however, the electrodes potential is slightly changing, and the anode reaches negative potential. If this is due to aging effects or variations from the cycling is yet to be determined.

This is something to further research and study because testing with PAT-cells could make testing on smaller scale easier to perform and also give a better insight into how aging affects the different electrodes.

5. Conclusion

The experiment in this thesis investigates fast charging, its effects on cell degradation and if adapting the current according to the varying cell capacity, leads to a slower aging process. The first protocol (FCP 1) uses current based on the cell capacity at the beginning-of-life (BOL) given by the supplier, while the second protocol (FCP 2) updates current based on cell capacity at given intervals. The cells used in the experiments were provided by Volvo Cars and are pouch cells with a Li-ion NMC cathode and a graphite anode.

This method is proposed to give an indication if adapted current should be implemented in battery management systems (BMS) and researched further. An adapted current should decrease in between cycles, lessening the strain on the battery, and thus promoting a healthier battery and a longer battery lifetime. If the degradation is shown to be very slow for FCP 1 then any attempt at adjusting current would yield only a small or non-existent improvement in battery lifetime. The method has been limited by available time, number of cells tested, a set constant temperature of 35 °C and one type of cell tested (pouch cell).

The results show a clear increase in battery lifetime of up to 340% by using FCP 2 compared to FCP 1. Capacity retention is 24-27% higher (depending on which cells are compared) for both cells (cell 3 and cell 4) fast charged according to FCP 2 at the end of cycling. The same trend is seen when evaluating the amount of energy throughput while charging. Charging time decreases a lot when fast charging according to FCP 1 while staying relatively constant when fast charging according to FCP 2.

A teardown is performed after 350 cycles on cell 2 and cell 3 where the cells show different amounts of degradation, strengthening the results. The main degradation of both battery cells is attributed to the consumption of electrolyte. For cell 2, most of the electrolyte dried out, while there are trace amounts left in cell 3. The SEM-analysis gives an indication of lithium plating for cell 2, fast charged according to FCP 1, but not for cell 3.

The second part of this thesis work is to construct and evaluate PAT-cells, where the electrodes are extracted from the same type of cell, as the commercial cells cycled. The method for the PAT-cells is to cycle them with the same FCPs used in commercial cell testing to be able to compare and validate testing on smaller scale. Unfortunately, the equipment does not work properly hence the cell cycling was cut short. The results show a degradation in capacity retention of 1-7% after 100 cycles. Since not enough cycles are performed the comparison as well as the validation of the equipment cannot be properly performed.

However, since the potential of the electrodes changes slightly during aging, it could indicate that SOC-based FCPs might need to be improved upon, not only by adapting the current but also the voltage level that the SOC threshold is based on.

It is our opinion that the results should be further tested due the potential this method has shown. Implementation of this protocol in BMS should be explored and if enactment of an updated capacity and current would prove to be too difficult then a simpler protocol could be constructed. This protocol could decrease current with a set rate to follow the same rate in which FCP 2 decreased by.

6. Future work

The topic of this study, i.e., how to optimize fast charging strategies to ensure longer durability of batteries, is constantly developing, and new information regarding research from fast-charging such as new fast-charging protocols, battery cell materials etc., is updated quite often. This is one of the reasons why this thesis work has been very intriguing and exciting. The tests performed gave an insight into what future development should strive for and what experiments could be done to further expand upon the results. Due to many limitations within this project, we have gathered some thoughts on how studies within this field could continue further.

One factor that limited this project is the time constraint of the thesis work. The battery cells could only be cycled for about 3 months. When cycling the batteries over a few years, the cycling could be adjusted to resemble the average driving and charging pattern of an EV-owner. After many years of cycling, a clear trend could be seen which would open up the option to do a derating function as previously mentioned in section 4.5. By doing this, a constant of how the average capacity is decreasing could be extracted which provides an easier way of implementing this into future battery cells. The derating constant would simplify the adjustment of the current, since the capacity would not need recurring calculations, but instead follow the same pattern throughout the cycling. Capacity checks that update the current may be difficult to implement in already existing batteries in EVs. At the same time there is a drawback with performing experiments over a long time since the development within this field is improving constantly and if waiting too long, the cells could be considered “outdated” when the experiment is finished.

Another limitation within this project is the temperature, it has been set to 35 °C which is done to increase the rate of aging in the cells. This does not reflect real life conditions where the cell temperature should be kept at 20-25 °C which is close to optimal operation temperatures. Since higher temperature leads to faster degradation, a lower temperature should slow it down. FCP 2 would then be able to derate current at smaller degradation intervals and thus increase performance even more. It might also show that FCP 1 doesn't degrade as much and the difference between them isn't as large. Further testing at normal operating temperatures is required.

Further, to perform the tests on many more cells than just two might be the most important part. When doing this, the FCPs can be performed on different types of cells as well since this project only uses pouch cells. Since the current is adjusted according to the capacity after 50 cycles in FCP 2, it can also be exciting to see how the capacity retention would look like if it is adjusted after fewer or more cycles, for example after every 20th or 100th cycle.

Since FCP 1 is the fast-charging protocol recommended by the supplier and used as a reference case, FCP 2 can be seen as the experimental one. Another suggestion for future work is to test out more, different fast-charging protocols. One suggestion that has been discussed during our thesis project is to lower the current with 1% after every 50th cycle. The results expected from that protocol is that the degradation effects should be in between FCP 1 and FCP 2. Since it is lowering the current, it is not putting as much strain on the battery in the later cycles, as FCP 1 does. However, since it is not considering the capacity of the battery while cycling, the cells should not have as long durability as when charging cells according to FCP 2. The tradeoff here would be that it could be easier to implement into current BMS.

Within this thesis project, the degradation phenomena have not been investigated in detail due to it being outside of the scope. This is also a suggestion for future work due to many possible degrading effects which could be mapped further with more investigation and research.

The work with EL-cells should be continued. It would be of great benefits to be able to perform testing with less material where the results can be used commercially. This would allow for greater sampling size since you cut down the cost of pouch cells and can invest it in testing equipment instead.

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