

Understanding the problem



University of  
Sheffield

**Finding Machine learning methods to model the  
degredadition of lithium based batteries under  
multi-stage CC charging**

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# 1 Introduction

There are currently over 7 billion people with access to electricity [1], the global consumption of energy is rising by  $4.5 \times 10^{16}$  Joules every year; 77% of which is from utilising non-renewable sources. With the affects of non-renewables on the environment still not fully realised, coupled with the concern of their finite-life nature, poses a constant need to increase usage of renewable sources. However, a large set of renewable generation methods fall under the category of variable renewable energy (VRE) sources [2], thus requiring a robust form of energy storage to solve the intermittent availability - methods of storage do exist and suitable solutions depend on the specifics and locations; as battery technology increases, the use of battery as a storage medium for the energy sector are ever more increasing. There is also the rapid increase in electric vehicle production, with a push to increase the adoption of EV's, 17.3 million cars were produced in 2024 [3] alone with a recent 20% average year-on increase. Lithium Ion batteries (LIB's) are the most common currently used batteries due to their desirable characteristics in energy density, aging properties, cost and more.

EV's and the energy sector account now account for 90% of the total lithium ion battery demand since 2016, the total lithium-ion battery demand is 10-times larger [4] since 2016. Different resources predict different forecasts of total battery growth [5, 6], regardless, it appears the rate of demand will not be decreasing in the near future.

LIB's, like all batteries and decrease in performance and capacity [7] until they are deemed unsuitable for their current use. This means eventually, all current batteries in use today will need to be disposed of, given that over 660,000 tonnes of earth metals were mined in 2023 alone, a strong case for slowing down the need for mining new materials is made, whilst keeping up with the demand (cite the iea thing). It's noted that battery chemistry is still a developing field, thus newer batteries may use less scarce materials, thus hard to truly predict the sustainability of mining these materials, for example, some reports (cite faraday) say cobalt supplies could be used entirely by 2040. Solutions do exist, recycling methods have been developed in order to extract the materials, termed battery metal recycling (BMR), however there are wildly different sources suggesting how much of current batteries actually are recycled (one showing 5%), (other showing 90%), and the technology to do so is still a developing field, there are many different methods (cite issurs), since most of the rare earth elements are within the batteries cathode, methods such as hydrometallurgical process can extract these, but at the cost of severe reduction in lithium yield (cite enji) - although there are

promising methods with some proposing a lithium recovery needing less than 40% the energy compared to mining the virgin material (cite enji yoo) whilst still recovering nickel and cobalt. There is also the rising interest in reusing the batteries before recycling, these batteries are referred to as second-life batteries (SLB's); in the EV use-case, the majority of the batteries are deemed finished once their usable capacity drops below 80% (cite), but often, due to the other parameters of the batteries needing to be of high standards for EV use, the 'dead' batteries can still provide a very useful service in less demanding applications, a common one being energy storage systems (ESS) [8], with claims reusing can reduce CO2 emissions by 56% as opposed to using natural fuel gas in situ. However there are again challenges associated, for safety there is the need for robust testing, sorting the batteries based on their current health states, non automated methods of extracting cells from their original pack. These additional steps mean the price of SLB's could become at a point where they are not much cheaper than new batteries, making the solution less attractive to many sectors [8].

In summary, there are methods that are developing to reduce the need for mining new materials and decreasing recycling energy, but ultimately, providing methods to increase the usable life of the current batteries serves as a direct way to reduce the impacts of batteries themselves and to meet their growing demand. If just 1 extra life cycle could be added to all EV's currently on the road (60 million (cite)), this could power all of the UK's energy needs for one day (cite gov.uk); as most batteries cycle life are well over 1000, any increment in cycle life can be greatly beneficial.

Much research has been done into both modeling LIB's and optimising their usage from both an energy and aging perspective.

Modeling and predicting how batteries degrade over time is pivotal for the growing use of lithium batteries as it allows informed decisions on how to best charge and use batteries to prolong life, as well allow optimisations on the charging throughout the batteries life.

## 1.1 Project Aims

This project aims first to characterise Lithium batteries behaviour by parameterisation, without ex-situ tests. This data will then be used in combination with work previously done to form the charging protocol based off minimising different objective functions. Data from this will be used to form the basis of a machine learning model in the hopes this model can, over time, allow

for a much simpler method of predicting a batteries future behaviour. If this model can show how different charging currents can affect degradation, a charging protocol which is derived from minimising such model will found and be tried in the labs to compare. If this is not achieved, atleast this project can validate existing predicted charging to minimise degradation will be tested, and the parameterisation of the batteries during degradation will be researched by using optimisation tools.

Figure x highlights the original idealised goal, to have a purely data driven model that can predict the output voltage over any cycle, thus been able to derive an idealised charging current to maximise the lifespan.

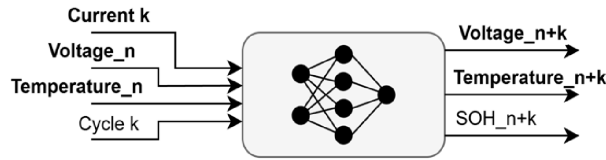


Figure 1: Initial end objective: Black Box Battery modeling degradation

### 1.1.1 Objectives

- Analyse ICLOCS2 and other methods in parameterising batteries under realtime use
- Modify previously developed charging methods for running in a lab to test actual results, inspect the affects of each method
- See how effective adapting the charging profile over the degradation is in increasing battery life
- Investigate ML methods to model the attained data to predict degradation
- Use the model to find a new charging method and validate

## 2 Literature Review

### 2.1 Lithium based battery degradation modes

**Notice:** Throughout the research and not coming from an electrochemical background, various literature denotes the meaning of *potential* and *overpotential* as slightly different given the context - there is likely equivalences, but of which the author of this report can't explain. For example cite(potential for confusion) shows a clear distinction between the common  $\phi$  symbol for potential as the *electrical potential*, however when talking about volts measured in the real world, the *potential difference* is usually the *electrochemical potential difference* (specifically of electrons), for two given points denoted by  $\Delta\tilde{\mu} = \Delta\mu + \Delta zF\phi$ . Overpotential in some instances, such as the Butler-Volmer model for current exchange, is a function of overpotential  $\eta$  and from cite greg is a difference in the current *electrical potential* and equilibrium potential, yet papers like S2352152X2400584X show overpotential more generally as the difference between any equilibrium potential and current potential. Thus confirmation should be made in regards to what type of potential and overpotential. Assumptions are given that potentials are a form of energy state, with a difference between two points causing a force, and overpotential as additional energy for an event to occur.

#### Basic Background of Lithium Ion Batteries

Lithium-ion based batteries are favorable largely due to the highly reductive nature of Lithium, taking the potential relative to hydrogen's reduction, lithium's half-reaction is  $-3.01V$  (cite gregg) relative to hydrogen's half-reaction, meaning lithium can release its outer shell electron relatively easily. Any cell's potential difference is the difference in potentials of the negative and positive solid state electrodes  $V_t = \phi_s^+(t) - \phi_s^-(t) - IR_{cc}$  where  $R_{cc}$  is the current collector or tabs ohmic resistance (cite liuying), so having negative half reduction of lithium, allows a variety of positive electrodes (where the oxidation occurs), to yield a high battery voltage. Coupled with Lithium's low molecular weight, a high energy density cell can be achieved to. The force produced by the potential difference between the electrodes drives the electrons via the external circuit, providing useful work, allowing lithium batteries to act as an energy storage medium.

Lithium batteries all follow the same basic principle of operation, as shown in .... They consist of a positive and negative electrode, electrolyte, a separator and current collectors. Unlike many

other battery chemistries, where the metal ions undergo chemical reactions at the electrodes which change the composition of the electrodes, lithium ions intercalate and deintercalate within the electrodes structure - they are in essence absorbed to and from the electrode structures (cite greg p). The negative electrode is usually of a graphite based structure, allowing up to 1 lithium atom to be stored per 6 carbon atoms, whilst the positive electrode can vary greatly between different lithium batteries; the most common are often composed of transitional metal oxides such as LCO, LMO, NMC, NCA (cite Alex K Hoech et al). The electrochemical state of lithium in the graphine is similar to that of individual lithium atoms, thus the electrode potential of the negative electrode is very similar to that of pure lithium (  $-3V$ ) (cite liquin), although its lithium storage abilities are usually lower than the positive electrodes. The lithium ions within the positive electrodes are in a much lower energy state, thus their reaction potential ranges from  $-1 - 2V$ , therefore, overall there is a large potential difference created by the want of the positive electrode gaining electrons and the lithium ions, and the want of the lithium within the carbon of the negative electrode to give up the lithium ions and electrons (relative to each other). In general, the negative electrodes used currently are carbon based, their capacity to hold the lithium (per unit volume) is greater than the positive electrodes used today as shown in figurexx (cite Alex), but as discussed later in this section the negative electrode is often the key limiting factor in degradation. The positive electrode materials vary greatly in material, but since this electrode has the biggest electrode potential, it has a largest impact on the overall cell voltage, and thus energy density of the cell, in addition since per volume it holds less lithium, electrode material with higher lithium capacity will be of greater affect than improving the negative electrodes capacity. The two reactions with lithium with the battery electrodes chosen for this project are shown below (during discharge, the reactions proceed from left to right, for the charging process, it's right to left):



The electrolyte is a medium in which the lithium ions can be transported via diffusion to the electrodes, the electrolyte does not however allow electrons to flow, only via an external circuit can they flow, . The separator acts as a structural barrier to prevent the opposite electrodes from touching, which would allow both electrons and ions to flow, causing major fire risks from the uncontrolled reactions. During the discharging process, a path for electrons is provided externally across the current collectors, (these collectors are not part of the reactions and are just a means to electrically

connect the electrolytes to a useful surface for soldering) the lithium stored within the negative electrode deintercalates, released as the positive lithium ion, whilst the electron leaves the electrode, traveling across the external circuit to the positive electrode, whereby the lithium ions meet the electrons. During charging, the applied voltage at the terminals will be above the difference of the electrode potentials, this applied overpotential causes the lithium ions within the positive electrode to deintercalate, traveling across the electrolyte to the negative electrode, where they intercalate within the graphite structure as the ions combine with electrons. The speed at which the ions flow is far slower than the electron transfer (cite greg), which contributes to the behaviour of lithium batteries.

Mention diffusion differences between charge and discharge (cite michael j lain)

### 2.1.1 Degredation methods

J'O. Kane et al. [9] summarises the key 3 modes of degradation caused within lithium ion batteries described below. Calendar aging is neglected in this discussion.

**Lithium plating** - Under certain charging conditions, the lithium ions within the electrolyte will join with the electrons outside the negative electrode, producing pure lithium metal - which can grow forming dendrites, depicted in figure xx. The most understood causes which accelerate this is when the negative electrode potential falls below that of lithium's own electrode potential potential [10, 11], becoming the most thermodynamically viable reaction. From electrochemical models, the potential at the electrodes is the sum of their open circuit potentials and overpotential. The overpotential is largely due to the kinetic overpotential at the interface of the electrodes, the Butler-Volmer equation shows as the current density  $j$  increases, overpotential  $\eta$ .

$$j = a_s i_0 \left[ \exp\left(\frac{\alpha_a F}{RT} \eta\right) - \exp\left(-\frac{\alpha_c F}{RT} \eta\right) \right]$$

Low temperatures and high SoC also increases Lithium plating, since it becomes harder for the lithium to intercalate within the carbon, the current effects are shown in figure xx in sections below. Modelling aims to characterise and capture potentials to help with controlling dendrite formation, methods from numerical electrochemical models like the p2D thing, using the butler-volmer kinetics to show key overpotentials, as well as state estimation approaches for realtime - however they all require many parameters about the specific batteries.



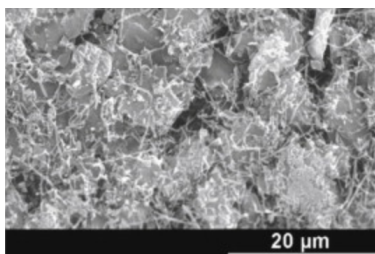


Figure 2: Lithium deposits shown in lighter grey on the graphite electrode [12]

**SEI Layer growth** - A layer known as solid electrolyte interphase is formed as soon as the electrolyte solution comes into contact with the negative electrode causing salts like  $\text{Li}_2\text{CO}_3$  to produce acids, followed by further reactions; this barrier acts to prevent electrons further reducing and using up the more of the electrolyte, whilst allowing passage of the lithium ions to intercalate [13]. If this SEI breaks apart, new SEI will form, taking more lithium up causing a reduction of lithium available for charging. Having the battery at very high and very low SoC can cause the SEI layer to thicken also [14]. SEI formation takes up electrons within its reaction, thus reducing battery capacity.

**Particle fracture** - The physical volume of the electrodes can change during the intercalation and deintercalation, this is a degradation feature that can't be avoided in order to charge and discharge a battery, some electrodes exhibit more contraction than others, silicon is significantly greater than graphite GREG. Concentration gradients, caused by high currents, within the electrode can also cause internal stress. This can overtime cause the electrodes to break down, either resulting in more SEI grown, reducing more lithium; inability for areas of lithium intercalation, reducing charge capacity; and separation from the binder causing either increased ohmic resistance or loss in capacity

In summary, high charging currents, extreme temperatures and extreme states of charge can accelerate degradation, thus to reduce this, charging and discharging at infinitesimally small rates and at optimal temperatures is best, however this neglects considerations such as charge times, thus a balance between these constraints should always be taken into account and the charging strategies used in this project attempts to do so.

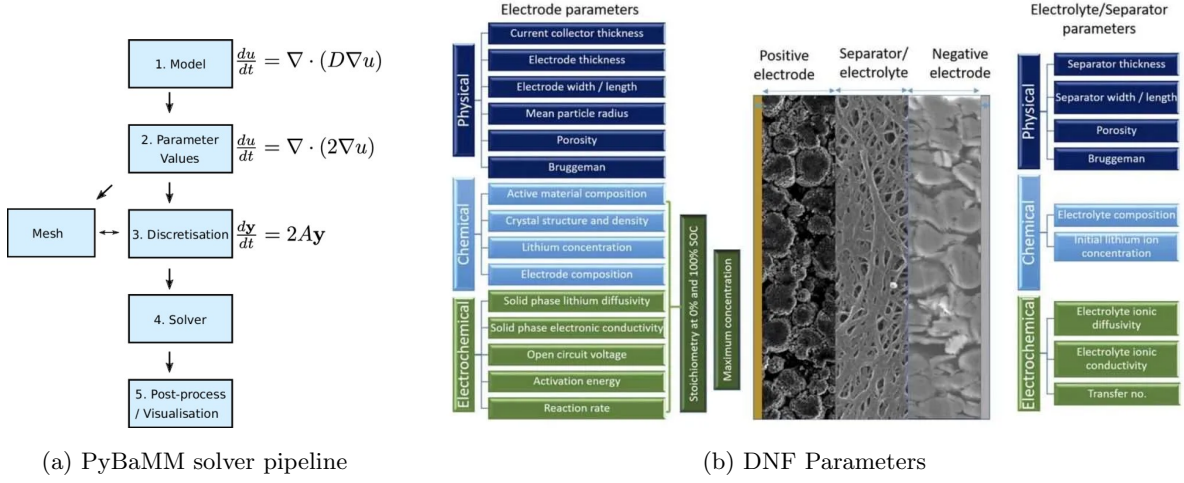


Figure 3: Parameters and solution steps for physics based modeling

## 2.2 Equivalent Circuit Model

There are many ways a lithium battery can be modeled, depending on the accuracy required, parameters available and computational power available. The two main categories are the equivalent circuit models and physics-based models (cite greg). The physics-based models integrate conservation laws as well as dynamical behaviour which leads to a set of PDE's with both scalar and gradient based boundary conditions. These require discretisation within the physical dimensions of the model to yield a set of ODE's which can be solved for, PyBaMM is a popular framework for such modeling and its process is shown in figure xx. Popular physics based models in order of computational complexity [15] are the Doyle-Fuller-Newman model [16], Single-Particle Model with electrolyte (SPMe) and SPM. The key problems of PBM is some require over 30 parameters to fully describe the properties and errors of such can accumulate over time, and depending on accuracy needed, the discretisation can yield hundred of ODEs (cite battery design).

Equivalent circuit models use standard lumped electrical elements to match the behaviour to that of a real lithium battery dynamics, it is this type of model most common in BMS systems today. T.Kalogiannis *et al* [17] provides common methods for obtaining the parameter values used in ECM's. The most common version is shown in figure xx, which is static in its dynamics. The key component is the  $V_{oc}$  ideal voltage source, this value becomes a function, (when neglecting temperature, degradation, charging-discharging hysteresis) of the batteries state of charge SoC,

given by

$$SoC(t) = z(t)/Q = (z(t_0) + \int_{t_0}^t \eta(\tau)i(\tau) d\tau)/Q$$

where  $Q$  is the batteries nominal maximum capacity and  $z$  is the current charge capacity. The  $R_0$  and  $R_1C_1$  branch are used to model the difference in terminal voltage compared to that of the  $V_{ocv}$  for a given instantaneous SoC, and  $\eta$  here is the colombic efficiency, usually 0.99 for lithium batteries, since some current will be used in irreversible chemical reactions within the cell cite gregg.

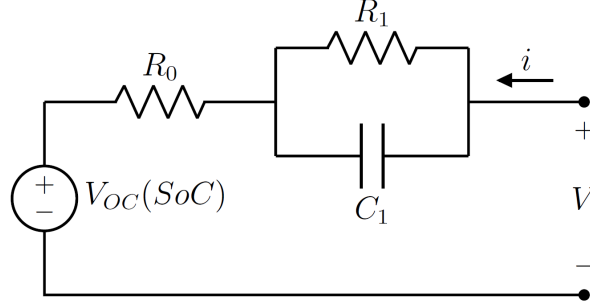


Figure 4: ECM model with 1st Order Polarizing dynamics

The common state space form of this system is given below

$$\begin{bmatrix} \dot{v}_1(t) \\ \dot{z}(t) \end{bmatrix} = \begin{bmatrix} -\frac{1}{R_1C_1} & 0 \\ 0 & 0 \end{bmatrix} \begin{bmatrix} v_1(t) \\ z(t) \end{bmatrix} + \begin{bmatrix} \frac{1}{C_1} \\ \frac{1}{Q} \end{bmatrix} i(t) \quad (3)$$

$$V_{batt}(t) = V_{oc}(z) + i(t)R_0 + v_1(t) \quad (4)$$

S2352152X2400584X provides the equation for the total overpotential within the battery derived from the Fuller-Newman electrochemical mode, here, the total overpotential  $\eta_{batt}$  is equal to the difference from the equilibrium battery potential  $U_{batt}$  and terminal voltage potential  $V_{batt}$ , i.e  $V_{batt} = U_{batt} - \eta_{batt}$ . This is analgous to the ECM model given above, whereby the overpotential (dynamical behaviour) is modeled by  $i(t)R_0 + v_1(t)$ , where the equilibrium potential is  $V_{oc}(z)$ .

$$\eta_{\text{batt}} = \left[ \begin{aligned} & \underbrace{\left( \Phi_{2,p}|_{x=L_p} - \Phi_{2,n}|_{x=0} \right)}_{\text{Electrolyte overpotential } (\eta_2)} \\ & + \underbrace{\left[ \left( U_p(c_{1,p}^s)|_{x=L_p} - U_p(\bar{c}_p)|_{x=L_p} \right) - \left( U_n(c_{1,n}^s)|_{x=0} - U_n(\bar{c}_n)|_{x=0} \right) \right]}_{\text{Li concentration overpotential } (\eta_1^c)} \\ & + \underbrace{\left( \eta_p^{ct}|_{x=L_p} - \eta_n^{ct}|_{x=0} \right)}_{\text{Kinetic overpotential } (\eta^{ct})} - \underbrace{R_f I_{\text{app}}}_{\text{Electrode ohmic overpotential } (\eta_1^\Omega)} \end{aligned} \right] \quad (5)$$

Whilst the voltage dynamics of lithium batteries do depend on temperature, this project keeps the ambient temperature to 30, thus realtime affects on the electrical dynamics are minimal. However modeling of the temperature is vital since temperature does directly affect degradation. cite the thermal paper. The irreversible is much easier to directly relate to the ecm, by basic ohms law and various levels of accuracy cant be obtained with different data avalaoable. However irreverisibility can not be directly infered from the ecm model. Cite dodgey paper, equated it to proportional to  $\partial U / \partial T$  where  $U$  is the OCV and  $T$  is the temperature, this value does change over the SoC of the battery and paper et all calculates the curve for a battery of similar to the test battery shown in figure x.x

$$mCp \frac{dT}{dt} = \underbrace{I^2 R_0 + IV_1}_{\text{irreversible}} - \underbrace{IT \frac{\partial U}{\partial T}}_{\text{reversible}} - \underbrace{hAT}_{\text{dissipation}} \quad (6)$$

A. Farman *et al* [18], shows that the OCV of lithium batteries can change throughout degradation, Lithium phosphate batteries showed a 20mV difference in regions of the SoC after aging of 500 cycles at 1C of charge and discharge. Additionally, while there exists methods of extracting an accurate OCV curve, it can take hours to accurately obtain to minimise overpotentials [17]. This is where numerical methods could be utilised to obtain the OCV curve and the other ECM parameters by fitting against a grey-box model of the dynamics, ICLOCS2 [19], will be used heavily throughout this project to see its ability to parameterise the battery dynamics. A.A. Mohamed et.al cite shows the many methods to obtain ECM parameters with methods ranging from analytical approaches based on various current inputs, to meta-heuristic optimization algorithms.

## 2.3 Charging methods

There exists many techniques to charge batteries with a graphical summary of the common methods shown in figure xx, cite Qian Lin highlights the different methods described below. The most common method is known as CC-CV charge. This is where the bulk charge is done such that the applied overpotential to the battery causes a constant current to flow, the voltage potential of the battery rises to a defined maximum point, the charging device then switches to a constant voltage charging mode, this fixed voltage is held until the charge current decays to a negligible or fixed amount - current still flows during this stage as there is still an overpotential between the batteries equilibrium state and the applied constant voltage. This is the simplest to implement without the need for an accurate SoC reading to risk overcharging (like would be with pure CC charging).

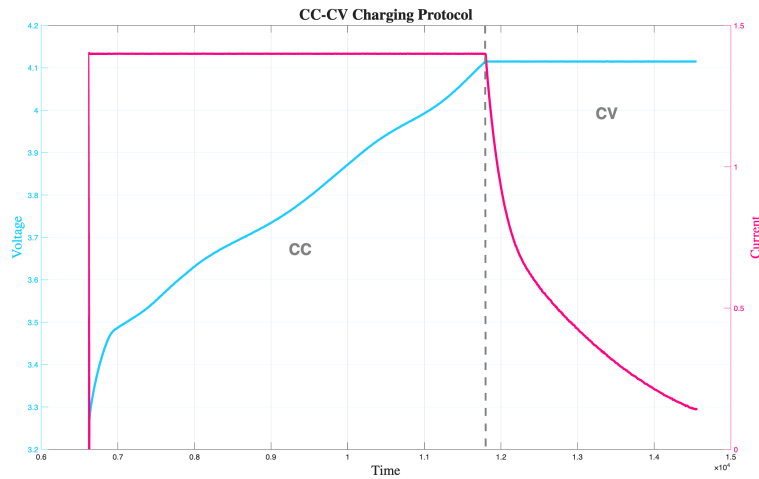


Figure 5: Charging segment extracted from the UoS dataset

CV is another method such that the voltage applied is the battery's maximum at full SoC, but since lithium batteries have low internal resistances, this can lead to a very high current draw at low SoC (where the equilibrium voltage is low), hence can cause accelerated degradation, cite quan mentions 40% capacity decrease after only 160 cycles. Boost charging provides a high current increase at low SoC, but only for brief periods and can significantly reduce degradation rate compared to CV.

AC charging is another method which aims to minimise the frequency dependant impedance of the battery, effectively allowing to charge the battery with a lower effective overpotentials, however as cite Qian mentions, there are disputes on the effectiveness of this method; coupled with the fact EIS (electrochemical impedance spectroscopy) is required to find the frequency points of interest to

target, this method wont be futher researched within the project.

Multistage CC is a popular method in reseach as it allows for the customisation of the charging throughout the cycle, this means various models can be used to optimise for a specific goal. Cite tucker for example, simulates various functions to minimise based of the ECM model provided in the section above. Electrochemical based models allows for a finer level of optimisation, such as ensuring the negative elecrode does not reach a potential to cause lithium plating, [20] models an observer to estimate the potentials, which could be used within optimisation as one of the constraints, figure xx shows their predicated potential voltages.

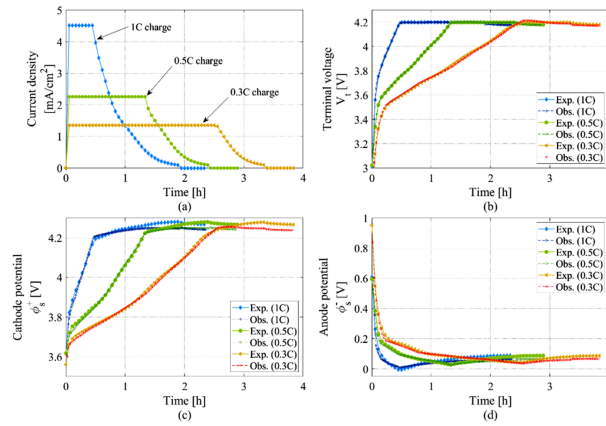


Figure 6: Estimated potentials in response to charge current

The multistage CC charging method provides a good framework to trial different charging methods, its balance of ease of implamentation and adjustability means it is this method which will be used in the project to optimise for degradedition.

## 2.4 Modelling degradation

There are three main categorical methods to predict the degradation of lithium ion battereies: Purley data driven methods, physics-based models and a hyrbdid (phuysics informed). F. Wang *et al* [21] highlitghts the advantages of the hybrid approach, stating issues and advantages with pure data-driven and pure physics model, they also present a means of categorising the common hybrid approaches.

Papers focusing on the physics based moedeling usually focus on a subset of degradedion modes, such as SEI thickness growth cite Mixed Mode Growth Model for the Solid Electrolyte Interface.

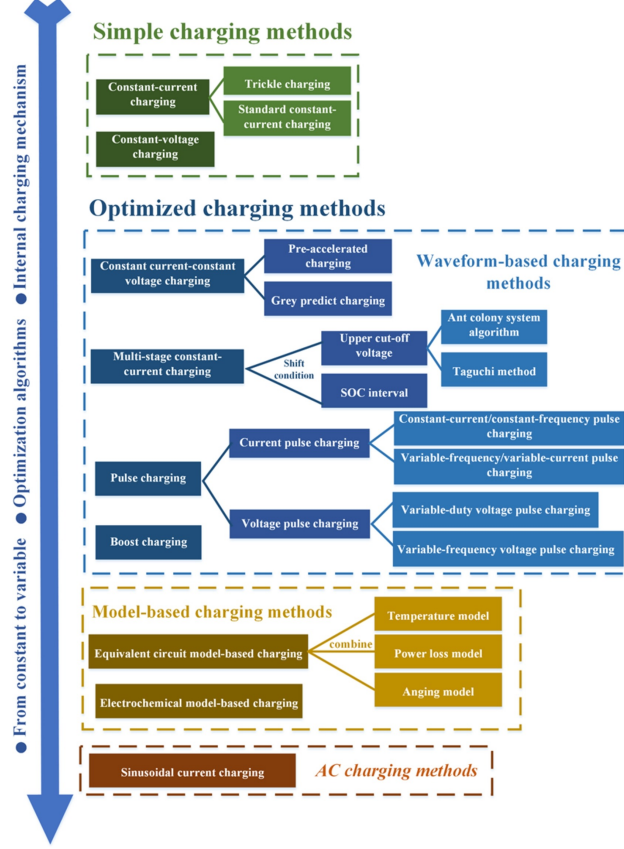


Figure 7: Common charge techniques, cite qian lin towards smarter

Most models start with the Doyle-Fuller-Newman model cite Dole Fuller to describe the transport of lithium within the battery (including potentials), then further sub-modules representing degradation mechanisms are integrated to yield the required models. For example s. okane *et al* [9] adds SEI, li particle cracking and lam as the degradation models. These approaches can model intricate parts within the battery, but require a lot of parameters about each battery under test, additionally, the discretised solutions are only ever as accurate as the models used. E j okane shows a more complete set of degradation modeling whilst Jishun applies ML to effectively learn the missing modes with good results.

Jishnu Ayyana by representing the electrochemical dynamics as  $\frac{dx}{dt} = f(\mathbf{x}, \mathbf{z}, \mathbf{u}, t; \alpha)$  and coupling to degradation dynamics  $\frac{dz}{dt} = f(\mathbf{z}, \mathbf{x}, \mathbf{u}, t; \theta)$  where  $\mathbf{x} \in \mathbb{R}^{n_1}$  and  $\mathbf{z} \in \mathbb{R}^{n_1}$  (once discretised), representing the battery electrochemical and degradation state variables respectively

Severson *et al* [22], provides one of the largest available dataset for battery charging. This paper takes uses a purely data driven approach in modeling, but uses engineered features such as the

change in discharge voltage curves between cycle 0 and cycle 100. Using a linear regression model, they predict the cycle to failure (defined when the capacity drops to 80% of original), it shows upto a 9.2% accuracy. Attia *et al* [12] takes the work of Severson *et al* to great use, using the model to predic how well differenct charge cycles last by cycling the batteries under random multistage CC cycles, predicting the degradedadition and using a bayesian optimisation strategy to find the next appropriate charging protocol - in a closed loop feedback shown in figure xx

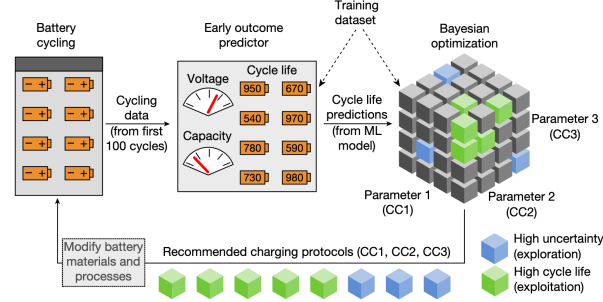


Figure 8: Closed loop workflow for finding optimal CC charging

### 3 Self Review

Choosing this final year project has, and will continue to, be a big personal undertaking. I knew nothing about batteries other than the very basics when starting, I also knew very little within systems & control other than simple LTI systems. Research has also been a struggle for me as many papers incorporate control methods or electrochemical background of which i dont have, a high amount of papers regarding the use of AI in modeling degradedadition are also behind paywalls.

A large amount of time has been spent of narrowing down the focus of the project, as well as preparing the opmitised controll desgins for the lab testing. As of 01/12/2025, the first two batteries are undergoing characterisation tests and the softwear side is nearly done. Focus will now be spent on researching more about the machine learning and ways to optimise charging from the modeling of the battery.



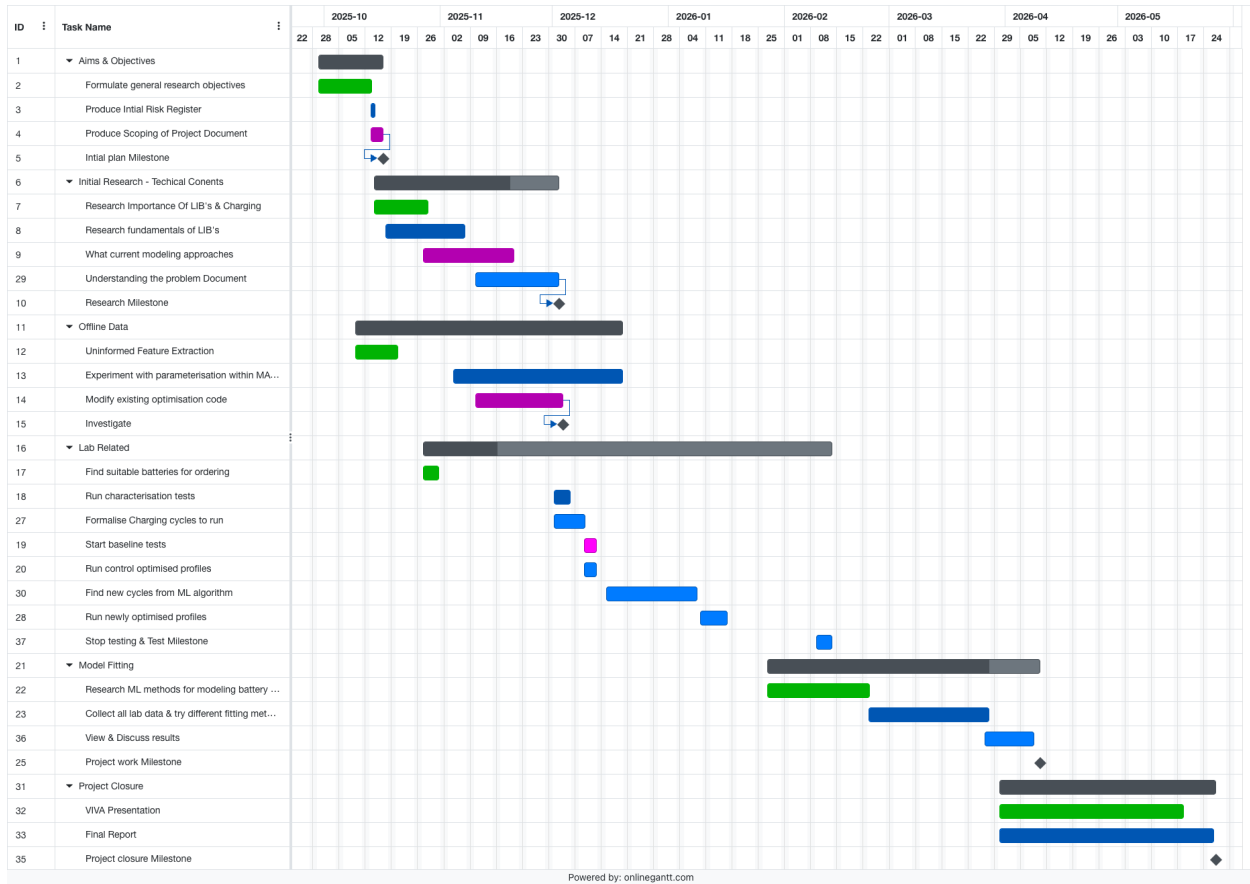


Figure 9: Gantt Chart of current project plan

**Note:** The following references may have duplicates and unused references, this should hopefully be fixed for the final report

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