

Understanding the problem



**University of  
Sheffield**

**Implamenting multistage constant-current charging  
methods in lithium based batterys to reduce  
degredation**

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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 widely 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 ISSUES), 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 in essence aims first to characterise any 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 (the level of affects the previously proposed objective functions will be also verified by acting these tests). 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. As will be discussed in the sections below, characterising batteries behaviour can require upto nearly 30 parameters about the specific battery in order to see the affects of degradation; even then, due to minor errors in soltuons can fail to predict future properties.

Figure x highlits the original idealised goal, to have a purley data driven model that can predict the output voltage over any cycle, thus been able to derrive an idealised charging current to maxamise the lifespan.

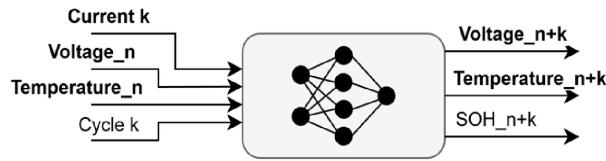


Figure 1: Inital end objective: Black Box Battery

### 1.1.1 Objectives

- Analyse ICLOCS2 and other methods in parameterising batteries during realtime use
- Modify previously developed charging methods for running in a lab to rest actual results, inspect the affects of each method
- See how affective adapting the charging profile over the degradation is in increasing battery life
- Propose and test new charging based on results of prevouls developed profiles
- Feed results to a data driven model to find a new optimal charging profile

## 2 Literature Review

### 2.1 Lithium based battery degradation modes

#### 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  $-3.01\text{V}$  (cite gregg), meaning lithium can release its outershell electron relatively easily. Any cell's potential difference is the difference in potentials of the negative and positive solid state electrodes ( $V_t = \phi_s^+(t) - \phi_s^-(t)$ , neglecting the battery terminal resistances), so having a very negative potential like the lithium reduction, 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 - they are in essence absorbed to and from the electrode structures (cite greg p), forming as loosely bonded lithium atoms within the electrodes. The electrodes for the negative side is usually 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 positive electrodes are often composed of transitional metal oxides such as LCO, LMO, NMC, NCA (cite Alex K Hoech et al). The lithium state in the graphite is similar to that of individual lithium atoms, thus the electrode potential of the negative electrode is very similar to that of pure lithium ( $-3\text{V}$ ) (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  $-1 - 2\text{V}$ , 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) Whiney well

### 2.1.1 Degredation methods

Simon et al. [9] summarises the key 5 modes of degradation caused within lithium ion batteries

**Lithium plating** - During discharge, the lithium ions intercalate into the positive electrode. Under certain charging conditions, the lithium ions within the electrolyte will join with the electrons outside the negative electrode, producing pure lithium metal - commonly referred to as dendrites, depicted in figure ... There is still ongoing research into finding the key causes and which factors speed up this process, but its most understood cause is when the negative electrode (carbon in the projects battery case) potential falls below that of lithium's own potential ([10, 11] Shichun Yang + Michael J. Lain). As previously mentioned, the lithiated carbon potential is slightly higher than that of pure lithium. From electrochemical models, the potential at the electrodes is the sum of their open circuit potentials and their overpotential, overpotentials are caused from the dynamics of the battery, i.e. during charge and discharge, thus, the overpotentials can drive the battery below that of lithium, as well as the SoC of the battery which increases the natural potentials (cite Zhiqiang). Meaning the more favourable reaction is to form lithium as opposed to intercalate. The overpotential is largely due to the kinetic overpotential at the interface of the electrolyte, SEI and electrode; it's shown that low temperature, high currents and high SoC can cause the negative potential to drive lower (cite Zhiqiang). 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 battery.

**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 [12]. 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 [13]. SEI formation takes up electrons within its reaction, and thus c

**Particle fracture** - The physical volume of the electrodes can change during the intercalation and deintercalation can cause



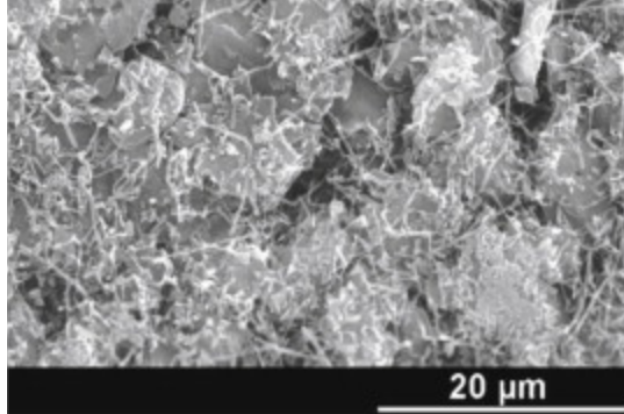


Figure 2: Lithium deposits shown in lighter grey on the graphite electrode [14]

Causes increased ageing and safety risks, it's the deposition of metallic lithium on the anode surface, happens at high charging currents and low temperature. Since during charging, the lithium ions move, through the SEI into the anode, if the ions cannot intercalate fast enough, they deposit and can become metallic lithium. Especially when charging is forced, local overpotential can cause the lithium plating, can cause dendrites

- SEI layer growth via pores  $\approx$  not really solvable, grows square root over time and cycle number
- Lithium plating
 

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- Active material loss (from parts mentioned above)
- SEI Breakages
 

Charging too **high** of a temperature causes mechanical stress on the SEI layer, causing it to crack and reform, consuming more lithium ions in the process. Loose SEI material can also float in the electrolyte, causing further issues.
- Electrolyte decomposition

## 2.2 Equivalent Circuit Model

This S2352152X2400584X nicely shows the overpotential of the battery relating to the difference of the output of the battery to the equilibrium potential of the battery. Shown as the summation of electrolyte overpotential, Li concentration overpotential, kinetic overpotential. Attempts to estimate the anode voltage based off reduced order electrochemical models and by use of a state estimator. In the electrochemical models, the anode potential is equal to the kinetic overpotential, anode equilibrium potential and the electrolyte potential, (cite) shows the solid-phase electrode potential is equal to  $U_{eq} + \mu_{ct} + \Phi_{electrolyte}$  th(cite).

and resistive overpotential. This are equated within the ecm model as the polarizing rc, and r. 0378775313020880 bib5 gives the

The

Whilst the voltage dynamics of lithium batteries do depend on temperature, this project keeps the ambient temperature to 30deg and moderately low temperatures, thus real-time effects are minimal. However modeling of the temperature is vital since temperature does directly affect degradation and will help the ICLOCS parameterisation. cite the thermal paper. The irreversible is much easier to directly relate to the ecm, by basic ohms law. However irreversibility can not be directly inferred from the ecm model. Cite dodgney 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 al calculates the curve for a battery of similar to the test battery shown in figure x.x

$$mCp \frac{dT}{dt} = \underbrace{I^2 R_0 + IV_1}_{irreversible} - \underbrace{IT \frac{\partial U}{\partial T}}_{reversible} - \underbrace{hAT}_{dissipation} \quad (3)$$

cite greg, shows methods of extracting the element values for the ecm, but the ocv curve can be particularly challenging, Especially given it hysteresis affects when charging and discharging. This is where iclocs and other models can come into play to help get these values

## 2.3 Charging methods

There was a paper comparing them in general, talk about the advantage of the different ones ect.

Talk about CC, MultiStage CC, ConstantPower, George Tuckers minimisation functions

## 2.4 Modelling degradation

Talk about how stevesons linear model uses the change in voltage overtime (first 100 cycles).

**Downside** - This requires a slow cycle to get the right features for the model, also only predicts EOL figure, useful for strong indication of battery prediction. However can not fully capture the input affects of current, only fitting from the affects of it. Additionally does not show how specific characyeristics change.

Attia takes the use of steverson to great use, using the model to predic how well differenct charge cycles last

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). Fujin Wang et al highlight the advantages of the hybrid approach, stating the main issue with pure data-driven and pure physics model, they also present a means of categorising the common hybrid approaches, given in figure x.

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. 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 degradedion mechanisms are intergrated to yeild the required models, the Dole Duller provides a set of coupled PDE's which when discritised with methods such as finite volume (cite pybam), serves as an approximation of the highly complex dynamics of the battery, others unclude the simpler single particle model and the single particle model with electrolyte SPM. For example simon e j okane adds sei, li paritcle crackiong anmd lam as the the degradedion models, this can be represented similar to Jishnu Ayyana by representing the electrochamiecal dynamics as  $\frac{d\mathbf{x}}{dt} = f(\mathbf{x}, \mathbf{z}, \mathbf{u}, t; \alpha)$  and couplining to degradation dynamics  $\frac{d\mathbf{z}}{dt} = f(\mathbf{z}, \mathbf{x}, \mathbf{u}, t; \theta)$  where  $\mathbf{x} \in \mathbb{R}^{n_1}$  and  $\mathbf{z} \in \mathbb{R}^{n_1}$  (once discritised), representing the battery electrochemical and degradedion state variables respectlvey. These approaches can model intericate parts within the battery, but require a lot of parameters about each battery under test, additionally, the discritised solutions are only ever as accurate as the models used. E j okane shows a more complete set of degradedion modeling whulst Jishun

applies ML to effectively learn the missing modes with good results.

Purely data-driven approaches have the ability to provide the most accurate predictions, however on their own suffer the problem that there is not currently a large enough set of data to train on - this is a problem as lithium batteries have multiple degradation modes as discussed, as well as a highly dynamical system when viewed as

Stevenson aims to

- Fmincon
- Greyest
- ICLOCS
- **2.4.1 Machine Learning**
- General Understanding
- LSTM
- 

### 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. Additionally I knew nothing about most machine learning, let alone modern neural networking. Research has also been a struggle for me as it is easy to follow references of references, as well as the disappointment when some research is shown that is either behind a paywall, or simply beyond my understanding.

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