

# *Investigation of Electrochemical Performance of NCA based Cathodes using Theoretical Analysis*

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A PROJECT REPORT  
SUBMITTED IN PARTIAL FULFILMENT  
OF THE REQUIREMENTS FOR THE AWARD OF THE  
OF

*Masters in Physics*

**Submitted by**

**Vaishali Dudeja**  
**2K20/MSCPHY/32**

**Pritesh Singh**  
**2K20/MSCPHY/22**

**Under the supervision of**

***Dr. Amrish K. Panwar***  
***(Assistant professor)***



***Department of Applied Physics***  
***Delhi Technology University***  
***(Formerly Delhi College of Engineering)***

***Bawana Road, Delhi-110042***

***May-2022***

## Declaration

We hereby certify that the work which is presented in the Major Project-II/Research work entitled **Investigation of Electrochemical Performance of NCA based Cathodes using Theoretical Analysis** in fulfilment of the requirement for the award of the Degree of Master in **Physics** and submitted to the **Department of Applied physics**, Delhi Technological University, Delhi is an authentic record of our own carried out during a period from **2021**, under the supervision of **Dr. Amrish K. Panwar**.

The matter presented in the report/thesis has not been submitted by us for the award of any other degree of this or any other Institute/University. The work has been published in SCI/SCI expanded/SSCI/Scopus indexed journal OR peer reviewed Scopus indexed conference with the following details:

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**Name (Roll no.):** Pritesh Singh, 2K20/MSCPHY/22, Vaishali Dudeja, 2K20/MSCPHY/32

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Phone: +91-9959870257

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*ENGINEERING DELHI TECHNOLOGICAL UNIVERSITY*  
*(Formerly Delhi College of Engineering)*  
*Bawana Road, Delhi-110042*

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*Vaishali*  
*Vaishali Dudeja*

*Pritesh Singh*  
*Pritesh Singh*

*DEPARTMENT OF APPLIED PHYSICS*  
*ENGINEERING DELHI TECHNOLOGICAL UNIVERSITY*  
*(Formerly Delhi College of Engineering)*  
*Bawana Road, Delhi-110042*

## *Abstract*

In this thesis, the modeling and theoretical simulations of Lithium-ion battery has been performed, for the improvement of its electrochemical properties and performances. The cathode material chosen for the simulation is  $\text{LiNi}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$  (NCA), along with graphitic  $\text{Li}_x\text{C}_6$  as anode material, and  $\text{LiPF}_6$  as liquid electrolyte. Initial simulation was carried out with variable thickness of cathode, and with charge discharge curve the optimized thickness was calculated. Thereafter, the discharge curve was plotted for 1st and 2000th cycle at three different cycle rate 0.5C, 1C, 1.5C for the optimized thickness, and potential drop due to SEI layer has also been calculated at these cycle rates.

The results obtained from the simulations shows consistency with the data reported in the previous literatures and experimental works. Hence the thickness of NCA cathode has been optimized and can be chosen as per required application and need. The results obtained from all three simulations show consistency with the previously reported literature based on experimental work. Hence, the cathode material (NCA) thickness is optimized and chosen easily according to the application and need.

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# Chapter-1

## Introduction

### 1.1 Background

Various non-renewable sources such as crude oil, coal, natural gases, etc. are being used now-a-days to generate energies. These sources pose a risk to the global atmosphere and climate change due to the production of CO<sub>2</sub> gases, air pollution, and greenhouse gases [1]. To overcome these issues, the research work is going on in this area to produce renewable energy sources such as solar cells, photovoltaic cells, micro-wind turbines, water energy, etc. [2]. Since, there is no regular source of energy, good energy storage devices are required by electric consumers for a constant supply, resulting in the supply which is not affected by the fluctuation. Now a days, to fulfill the constant supply demand, rechargeable batteries are a useful energy storage alternate.

Air contamination produced by the non-renewable energy source vehicle can lessen by supplanting these vehicles with an electric or half breed electric vehicle. Optional capacity gadgets are probably the best arrangement. In an electric vehicle, a powerful thickness battery furnishes the capacity to infer with greatest speed as well as to speed up the vehicle and jump on a slope anyway the battery with high energy thickness gives most extreme distance capacity to the vehicle. Regardless of the progressions in energy age, the energy area is as yet attempting to get a solid, high energy and power thickness, convenient and versatile energy stockpiling in the expected structure, from little scopes like smartwatches or pacemaker to enormous scopes like electric/mixture electric vehicle and burden evening out application.

### 1.2 Cell/Battery

Rechargeable battery is an electrochemical cell. It has a cathode (a positive electrode) and anode (a negative electrode). Both of them are separated by a solid separator and filled with liquid electrolyte and follow the multiple charge – discharge.

There are many rechargeable batteries, now-a-days, lithium-ion battery (LIB) is the convenient technology. These batteries have power density and energy and their cycle capabilities is also better than other types of rechargeable batteries. To maintain the quality is important in the modern world and its applications like electrical and hybrid vehicles and another important sectors i.e., energy storage systems are used in renewable energy applications. The lithium-ion batteries follows the principle i.e. during discharge process, lithium ions move towards the positive end (cathode) through the organic electrolytic medium from the negative end (anode). Mostly, the anodes in lithium-ion batteries are made of graphite.

#### 1.2.1 Types of batteries

There are two main classifications of batteries:

1. Depend on their electrochemical reversibility
  - a. Primary Battery
  - b. Secondary Battery

## Primary Battery

Batteries that can be recharged as their electrochemical regenerative reactions are called Primary Batteries. These batteries have different sizes such as D, C, AA, AAA, PP3 and coin cell. Used where charging is not possible. Primary batteries remain relatively high and low specific energy, often used by devices that use a small amount of power to allow the battery to last as long as possible.

## Secondary Batteries

The batteries can be recharged as their electrochemical reaction is reversed using a positive external voltage in the reverse direction of the current discharge battery. Second batteries are used when there is a need for high drain applications, the use of a single charged battery is expensive or impossible. A second battery for small capacity battery used on portable devices such as wireless phones, laptops, mobile phones. Secondary batteries for high power consumption in hybrid or electric vehicles and load generation to control power supply. Ni-Cd battery, Lead-acid battery, lithium-ion battery etc are the examples of secondary batteries.



**Ni-Cd Battery**



**Li-ion Battery**

**Fig. 1 a) Nickel-Cd Battery, b) Li-ion Battery**

**Table1- The comparison of different types of batteries**

Batteries	Li-ion	Pb -acid	Ni-Cd
Lifetime/cycle	400~1200	200~500	2000
Working Potential (V)	3.6/3.85	1.0	1.2
Gravimetric energy density or Specific energy (Wh/Kg)	100 -265	40-60	40-60
Volumetric Energy density or energy density (Wh/L)	250-693	100	50-150
Specific Power (W/Kg)	250-340	90-120	150
Self-Discharge (% per month)	2.5-5%	3-5	10
Charge/ Discharge efficiency (%)	80-90	90-99	70-90
Weight	Light	Heavy	Heavy

## 1.3 Lithium-ion Batteries

The lightest metallic component in the periodic table, most noteworthy electrochemical potential and the most elevated specific energy (Wh/Kg) make lithium as one of valuable material in electrochemical examination. Despite the fact that, lithium metal as an anode in the battery-powered battery gave exceptionally high energy thickness, undesirable dendrites development on the anode during cycling turned into a major issue with such batteries.

Currently lithium-ion battery is one of the most popular technologies of the battery sector. Lithium-ion batteries are better in cycle capabilities, energy and power density. These points are a major requirement in the modern world. Few of the applications like electrical and hybrid vehicles and another important sector i.e., energy storage systems are the good examples. All types of lithium-ion batteries have the same Chemistry; at the time of discharge, through the organic electrolyte, ions of lithium move from anode to cathode. The anodes in most of the technologies use graphite. The differences are among the cathode materials which contain nickel, manganese, or cobalt in different proportions. Different cathode types allow high intercalation levels and lithium insertion which ultimately produces large energy storage quantities. Solid Electrolyte Interface (SEI) layer may be defined as a layer formed on both ends of the anode. SEI is electrical insulation and leads to the potential drop but has enough conductivity of ions so that the battery may function normally. the SEI layer protects the battery but it reduces the capacity. In case of absence of SEI, Li-ion might not get that much long life which it possesses.

### Advantages of Lithium-ion batteries:

- It has high- (load capabilities with Power Cells, specific energy)
- Their cycle is long and shelf-life is extended
- They are free to maintain
- These batteries have low internal resistance and high capacity
- The coulombic efficiency is good

### Disadvantages of Lithium-ion batteries:

- To prevent thermal runaway, it requires protection circuit
- With high voltage and high temperature, li-ion batteries may break
- At temperatures below 0°C, fast charging is impossible (<03°C, <32°F)
- When these batteries are shipped in larger quantities, proper transportation regulations are required

## 1.4 Cathode

An oxidizing electrode that takes electrons from the current i.e. external and at the time of the reaction i.e. electrochemical, it gets reduced.

### **Following properties are possessed by Cathode materials:**

- Low cost
- Oxidizing agent are efficient.
- working voltage are useful
- Ease of fabrication
- Stable when in contact with electrolyte

There are different types of cathode materials such as layered,  $\text{LiCoO}_2$ , spinel,  $\text{LiMn}_2\text{O}_4$  (LMO), olivine,  $\text{LiFePO}_4$  (LFP) and other compounds of similar structure [3,4,5]

## **1.5 Lithium Nickel Cobalt Aluminum Oxide (NCA)**

Among the all types of developed cathode materials, the layered materials containing cobalt, nickel, and aluminum with different concentrations are of good capacity, good energy and power densities. High intercalation levels and lithium insertion are allowed by different cathodes which leads to large energy storage quantities. The lithium nickel cobalt aluminum oxides (NCAs) with variable composition of Ni, Co and Al content are a group of mixed metal oxides [6]. Due to their application in lithium-ion batteries, some of them become important. NCAs are used as Basic substance on the positive electrode (cathode). The empirical formula is  $\text{LiNi}_x\text{Co}_y\text{Al}_z\text{O}_2$  with  $x + y + z = 1$ . The rechargeable lithium-ion batteries consisting of NCA cathodes are used in the electric vehicles (EVs) and electric appliances with  $x \approx 0.8$ . They are the one currently available in the market. The batteries with high Ni content in NCA cathode are known as Ni-rich batteries. 3.8 V to 4.1 V is the range for the voltage of those batteries, and nominal voltage is between 3.6 V to 3.7 V. Hence,  $\text{LiNi}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$  (NCA) i.e., Ni-rich layered lithium transition-metal oxides are archetypal cathodes used for EV batteries. Because of their low cost, high reversible capacity and good rate capability. These materials have been studied extensively during the last two decades [7]. It has many applications such as in electric cars, power grid and medical devices. One of the leading manufacturers of electric vehicles like Tesla, uses NCA batteries in their electric vehicle's system.

Energy Storage is same in NCA as well as NMC batteries. Though NCA batteries are not widely used, but are used in the electric car industry and is quite promising. The usable charge storage capacity of NCA is about 180 to 200 mAh/g, which is less than the theoretical storage capacity, 279 mAh/g [8].

However, the capacity of NCA is much higher than other types of alternative materials of similar layered structure such as lithium iron phosphate  $\text{LiFePO}_4$  (LFP) with 165 mAh/g,  $\text{LiNi}_{0.33}\text{Mn}_{0.33}\text{Co}_{0.33}\text{O}_2$  (NMC 333) with 170 mAh/g and lithium cobalt oxide  $\text{LiCoO}_2$  (LCO) with 148 mAh/g. NCA enables batteries with high energy density because of high voltage. Another benefit of NCA is excellent High specific energy, fast charging capability, long life span and good specific power. However, since there is need of extra safety features and circuitry, NCA materials also have demerits in terms of safety issues.

Hence, the theoretical studies in lithium ion-battery have been attempted on NCA based cathode material. Therefore, using the simulation with defined initial parameters, a model has been prepared. Using the variation of the thickness of the NCA cathode, a one-dimensional (1-D) model is prepared. Finally, for the best electrochemical performance the optimized thickness was estimated and polarization effect, charge-discharge voltage etc. were investigated.

### **Advantages of NCA:**

- Specific energy of NCA is high
- It has reasonably specific power
- Long life span of NCA is high

### **Disadvantages of NCA:**

- It is less safe than Lithium-ion batteries, as it requires more safety features while using in electric cars
- It is expensive as compared to other Lithium-ion batteries

*Theoretical studies have been done on the battery with NCA as cathode material. For this, a model was being prepared, and some initial parameters were set and defined. The model prepared was a 1D model, where the thickness may vary (of the cathode). Initial parameters have also been written in the table below.*

## Chapter-2

### Literature Review

**Zhang, Sheng Shui. 2007** reviewed the different types of separators used in the liquid electrolyte of a lithium-ion battery. Divide the battery divider into three groups depending on the structure and composition: (a) Microporous Polymer membrane (b) non-woven fabric mats and (c) inorganic composite membranes. All three divider components have different properties or properties such as Microporous Polymer membrane with a thin body and thermal insulation properties, untreated fabric is characterized by increased porosity and low cost but in composite membranes it has outstanding moisture properties and exceptional thermal stability. In this review, the production of separators, features, performance, and modifications discusses the safety and performance of batteries. In this review, he concludes that the separator should be thin and have strong holes due to high power and battery power, but dangerously reduce safety and cycle life as it reduces battery power.[9]

**Xiao, Xinran, Wu, Wei and Huang, Xiaosong. 2010** reviewed the pressure inside the battery while charging and charging. The separator is a perforated membrane placed between direct or indirect electrodes to prevent physical contact between them and the flow of an electronic charger while allowing ionic transport. Due to lithiation and de-lithiation, there is a change in the electrode size and a difference in thermal conductivity between the two battery components that may cause pressure on the separator. According to this review, to check the pressure inside the battery, there is no way available until then. Therefore, the differential pressure analysis of the cell is performed in a limited multi-scale element. In a separator, a small dent is natural due to the electrode particles under pressure. The surface roughness is higher in curved areas compared to the typical divider areas. Therefore, greater difficulty is found in the confined area of the separator. Particle size, packing, and battery cell size determine the stress state and its size in the separator. There is a strong pressure in the separator if the pattern of loose particles while in the case of a closed particle pattern, under normal pressure and sufficient pressure in the separator, there is a compressive pressure.[10]

**Cai, Long and Ralph E. White. 2011** Studied the thermal conductivity of a lithium-ion battery during the output of a constant power with a pulse with and without pulse electrode. For this purpose, remove the battery first 3000s at  $C / 2$  and then  $3C$  until the voltage drops to 2.5V. He concluded that the cell is in a better temperature-separated state, i.e., in the adiabatic state it has a greater excretion rate compared to the isothermal state. The reason for this is that the cell temperature rises very rapidly with an output value of 1 C because the diffusion coefficient of the binary electrode is high which reduces the diffusion limits under adiabatic conditions than isothermal conditions. He also concluded that as the cells heat up as the output power rises when the collapse of the cell may occur due to hot temperatures.[11]



**Satish Kumar B. Chikkannanavar 2014** reviewed a summary of the emerging cathode material, which is as yet being created for use in electric vehicles. An unique combination, coordinated cathode frameworks offer benefits over a cathode made of a single material. The advantages are, longer cycle life, and low cost. In this manner, lithium-particle batteries that utilization spinel as a cathode item might be especially difficult in auto impetus application applications where battery duration longer than different working temperatures is significant. Spatial changes in accordance with further develop cathode molecule strength and respectability and to further develop cell wellbeing execution have been utilized effectively in numerous cathode frameworks and will probably stay a part of single-part or incorporated frameworks. Progress in growing better approaches to further develop strength, wellbeing, and security will proceed and may create ceaseless improvement. Over the long haul, new synthetics will be expected to meet the energy needs of PHEV and BEV applications. Additionally, quick charging turns out to be more productive to address the issues of the advanced way of life. Coordinated frameworks can shape the centre of the parent cathode which is especially equipped for accusing of top calibre. Right now, Li-rich mixtures and Li-sulphur frameworks have extraordinary guarantee, yet halfway advances utilizing a portion of the methods depicted here might be advantageous and examples found out about collecting and covering cathode frameworks will probably work. Otherwise, advances in production may lead to less expensive cathode components. Cathode systems for assembling and covering may work. Otherwise, advances in production may lead to less expensive cathode components.[12]

**Ruhul Amin 2015** reviewed  $\text{LiNi}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$  (NCA) Electronic and Ionic Transport in detail. Regardless of many businesses utilization of NCA as a good anode for Li-ion batteries, and its long examination history, its significant vehicle properties are not surely known. These designs are fundamental for planning high-thickness Li-ion batteries. With the help of extraction polarisation, impedance spectroscopy, and dc polarisation the transport properties of NCA are researched. A striking outcome is that the lithium ionic diffusivity rivals  $x$  appearance an angular bend with at least  $x = 0.5$ , while the cell unit boundaries show the contrary pattern. This proposes that cation arrangement greatly affects the development of the design than in the Li layer division, as opposed to other layer oxides. From temperature-subordinate estimations in electron-hindering cells, the beginning strength of lithium-particle conductivity (diffusivity) is gotten at 1.25 eV (1.20 eV). During the electrochemical process the use of chemicals is very much limited in the transportation of lithium ions, but it is more than enough faster throughout the charging range to allow charging / discharge of the micron scale particles at effective Cycle rate values.[13]

**Rahul Deb Pal, 2015** In this paper the creators streamlined a couple of boundaries of the battery model and approved the outcomes with the charge-release bend by exploratory outcome. Their exploration studies are useful to observe the condition at which the battery utilization is greatest. In this trial, the battery charge and releases multiple times with a sluggish pace of 0.04C at room temperature to accomplish a steady limit. Followed by galvanostatic charging and releasing at a 1C rate with voltage range 2.8 V-3.8V at 10°C, 30°C, and 40°C. Contrast these outcomes and the full-cell model of the one-

layered isothermal lithium-particle battery model accessible in COMSOL Multiphysics 5.0. This might utilize used in planning electronic gadgets and the board frameworks of battery.[14]

**Florian Hall 2016** reviewed that there is asymmetry in Discharge/Charge Curves of Lithium-Ion Battery. The NCA shows an alternate asymmetry in release/charging conduct to mass stoichiometry. He points out the essential electrochemical relations, to be specific, the Nernst condition and the Butler Volmer condition, had the option to deliver this conduct when a two-venture response was taken. The two-venture strategy comprises of (a) lithium-particle adsorption from electrolyte to the dynamic particles under electron move, and (b) the mix of lithium adsorbed iotas in the mass region. The asymmetry of the NCA anode half-cell test information turned at 0.1 C-rate can be duplicated in mass with this basic model. The model parameters show two other solutions, which predict the full (highly covered) area or the total area of lithium mass.[15]

**Frans Edison 2017** reviewed the State of Energy (SOE) of LiNiCoAlO<sub>2</sub> battery pack considering Cell Balance and Energy Efficiency. Further developed the power measurement (SOE) of the battery pack while considering the inconsistent voltage among cells and energy proficiency utilizing power calculator and vector support (SVM). The power calculation is done by gathering the approaching and active force of the battery pack while a charging. The power is gathered after some time and contrasted with the asserted power with decide the SOE of the battery module. The test was completed in two stages using NCA batteries with an ostensible voltage of 3.6 V. In the primary test, power estimations were performed on a solitary cell to decide the cell qualities of every battery in light of C5, C10, and C20 yield techniques. The consequences of the investigation show that the productivity of the power in the charging and release process for the result rate was  $0.8974 \times C^{(\exp(-0.033))}$ . Data set from single battery cell parts are coordinated into a survey table that shows the connection between SOE, current and voltage of the battery. The perception table was utilized as the preparation informational indexes for SVM to create a battery cell model and SOE module. A subsequent test was performed to gauge the SOE of a battery module comprising of 10 batteries in a series association in a similar result as a solitary cell test. The consequences of the SOE estimation utilizing an outspread base vector relapse model, with an expense viability of 30, and epsilon worth of 0.04, and a piece worth of 1 portion, give the r.m.s blunder esteem - a error of less than 5% indicating acceptable result. Since there will be imbalances in the cell y during the charging and discharge process of the battery, the minimum SOE value is taken as the reference value for other cycles.[16]

**Dao Yong Wan 2018** reviewed NCA cathode materials induced by Mn and Ti to work on electrochemical structures. Prepared NCA for hydrothermal reaction at 170° C. Translucent glasslike stages and morphologies of different lithium nickel cobalt aluminum oxide cathode components are described by FE-SEM, XRD, and molecule size examination. Te CV and galvanostatic charge/release tests are utilized to recognize the electrochemical properties of cathode materials. Titanium and Manganese doping caused an expansion in cell volume. Enormous Tis volume likewise worked on the electrochemical properties of cathode materials because of the way that Mn<sup>4+</sup> and Ti<sup>4+</sup> were brought into the octahedral grid taken by Li-particles to build the Li layer space and, therefore, further developed

lithium dissemination energy. Subsequently, the NCA-Ti terminal showed a high result limit of 179.6 mAh/g after the primary cycle, around 23 mAh/g higher than that got by the opened NCA anode, and 166.7 mAh/g after 30 cycles. The NCA-Ti electrode is detected the efficiency of 88.6% on statistics on initial charging and output power. Additionally, the NCA-Ti cathode material showed the best stability of 93% cycles up to 30 cycles.[17]

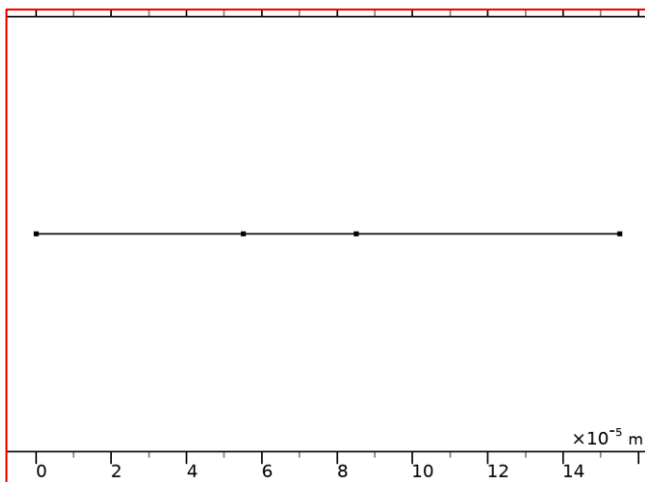
**Karina Diah Rosa Ekawati 2018** reviewed comparative study of NCA Cathode Material Synthesis Methods of their Structure Characters. Investigated the impact of the various mixing methods available to obtain NCA assets with better brands. NCA powder is acquired from direct, hydroxide co-precipitation synthesis, carbonate co-precipitation, , co-precipitation and solid state synthesis, characterized as NCA-DC, NCA-CC, NCA-HC, and NCA-SS separately, which is visualized using an X-ray diffractometer. . A structural study conducted comparing the results with commercial NCA XRD patterns also confirmed the identification of XRD patterns samples in JCPDS 871562. Based on the structural analysis, all samples showed a well-structured hexagonal structure. From the lattice parameter study, he concluded that the best method was developed with a sample obtained from direct coefficient heat.[18]

**Gyeong Won Nam 2019** reviewed volume Ni-Rich NCA Cathodes Blurring: The Effect of Extra Microcracking. He assembled the NCA to research the instrument of the Ni-rich NCA cathodes. Limit maintenance and warm properties of the cathode fall apart as their result limit increments when the Ni segment is expanded. The decrease in portion is decidedly corresponded with the distinction in anisotropic volume brought about by H2 – H3 stage change and the subsequent pace of minor breaks. Albeit every one of the three cathodes start to create microcracks in the wake of being charged at 3.9 V, the limit at which microcracks are conveyed without particles is diminished with the expansion in Ni content. These microcracks subvert the trustworthiness of the cathode hardware and work with the infiltration of electrolyte into the molecule centre, which speeds up the corruption of the surface region of the principal particles. Therefore, reducing or delaying H2 – H3 conversion is key to improving the performance of Ni-rich NCA cathodes.[19]

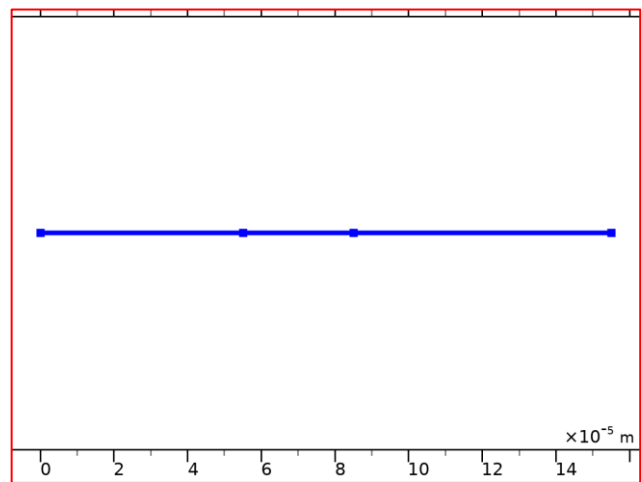
## Chapter-3

### Model Development and Simulation

COMSOL Multiphysics is software or formerly calls a finite element modelling program for simulation mostly in scientific research, design, and development of different devices and engineering fields. COMSOL Multiphysics software uses ordinary or partial differential equations (PDEs) to provide the platform for modelling any physical phenomena. This software can run or install nearly all platforms such as Linux, Unix, Mac, and all versions of Windows. A model of 1D Lithium-ion battery using COMSOL Multiphysics software developed for simulation.



**Fig.2: 1-D model geometry of battery without electrolyte**



**Fig.3: 1-D model geometry of battery with electrolyte**

Figure 2 designates the 1-Dimension model geometry which is also used in the models prepared for lithium-ion battery without electrolyte. Here, the length from 0 to 5.5 is for the negative electrode i.e., Anode build of Graphitic Electrode,  $\text{Li}_x\text{C}_6$  material.

Between 5.5 and 8.5, the geometry indicates the length of the separator and from 8.5 to 15.5 is the length of the Cathode material used as  $\text{LiNi}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$  (NCA).

Figure 3 shows the whole cell geometry after dipped into the liquid Electrolyte,  $\text{LiPF}_6$  and it is shown by blue color.

In this model, the thickness of the anode is kept fixed as  $55\mu\text{m}$  and for the separator, it is  $30\mu\text{m}$ , respectively [20] and the thickness of the cathode is varied. The defined values and the initial parameters are used for the simulation as separator, anode, cathode and other parameters may be seen in table 2.

# Chapter-4

## Parameters

Description	Value	Anode	Cathode	Separator
Particle radius negative electrode	-	2.5E-6 m	-	-
Particle radius positive electrode	-	-	2.5E-7 m	-
Solid-phase volume fraction positive electrode	-	-	0.42	-
Electrolyte phase volume fraction positive electrode	-	-	0.41	-
Bruggeman coefficient for tortuosity in the positive electrode	-	-	2.98	-
Solid-phase volume fraction negative electrode	-	0.384	-	-
Electrolyte phase volume fraction negative electrode	-	0.444	-	-
Electrolyte phase volume fraction separator	-	-	-	0.37
Bruggeman coefficient for tortuosity in separator	-	-	-	3.15
Length of the negative electrode	-	5.5E-5 m	-	-
Length of separator	-	-	-	3E-5 m
Length of the positive electrode	-	-	7E-5 m	-
Cell temperature	318.15 K	-	-	-
SEI layer conductivity	5E-6 S/m	-	-	-
Initial SEI layer thickness	1E-9 m	-	-	-
Number of cycles	2000	-	-	-

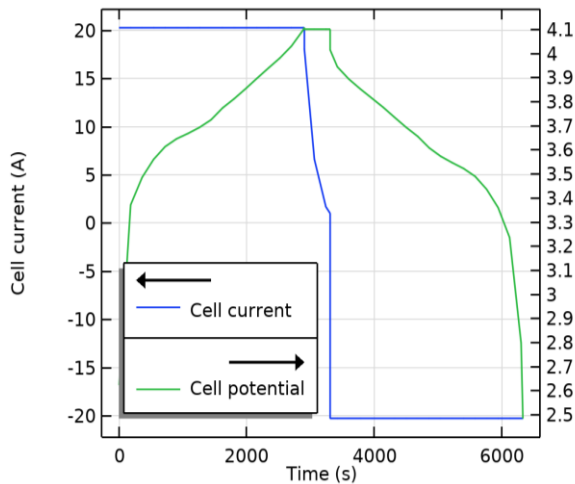
**Table 2: Initial Parameters of the model**

The 1D model of the battery is predefined with the initial parameters, and these parameters have been set or defined from the known parameters defined in the literature and some adjustable. Particle size of cathode, particle size of electrode has been kept constant. SEI layer has been given a initial thickness, and the cell is operated at a constant temperature higher than room temperature. The length of separator and anode has been kept constant, and length of cathode has been given a initial thickness and then varied with increment of 10 $\mu$ m, from 50 $\mu$ m to 100 $\mu$ m thickness.

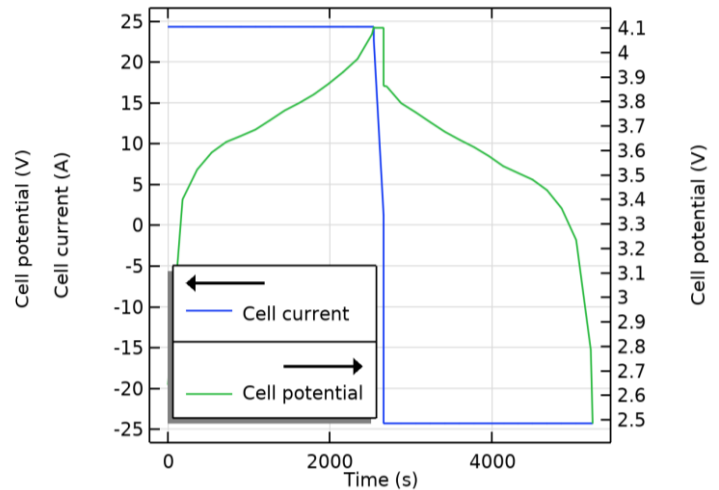
# Chapter-5

## Results and Discussions

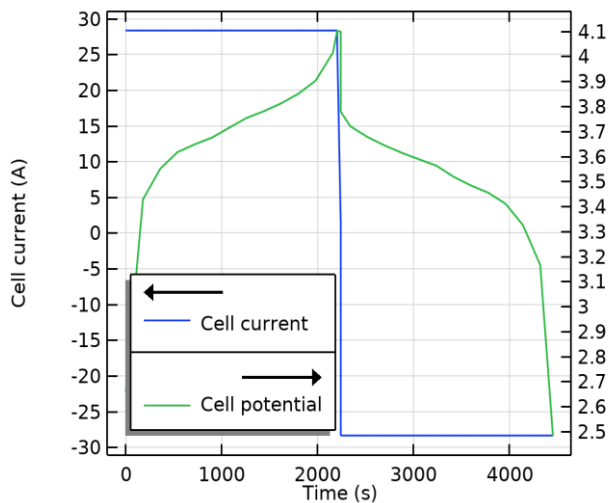
The below graphs shows the charge and discharge curve of the battery at the different thickness of NCA cathode. Fig.4 represents the charge and discharge curve for the thickness of 50 $\mu\text{m}$ , fig.5 shows the same for 60 $\mu\text{m}$  thickness of NCA cathode. The thickness has been increased to the 100 $\mu\text{m}$ , with increment of 10 $\mu\text{m}$ , and their charge discharge curves is shown in the fig.6 for 70 $\mu\text{m}$  thickness, in fig.7 for 80 $\mu\text{m}$  thickness, in fig.8 for 90 $\mu\text{m}$  thickness and in fig.9 for 100 $\mu\text{m}$  thickness.



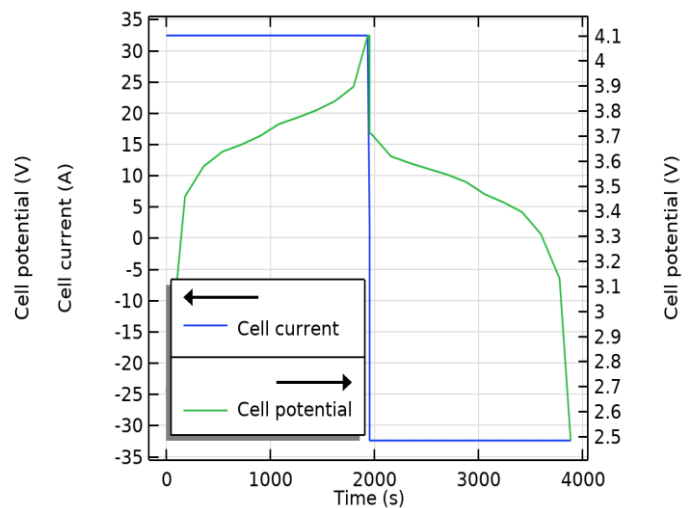
**Fig.4: load cycle at 50 $\mu\text{m}$  thickness**



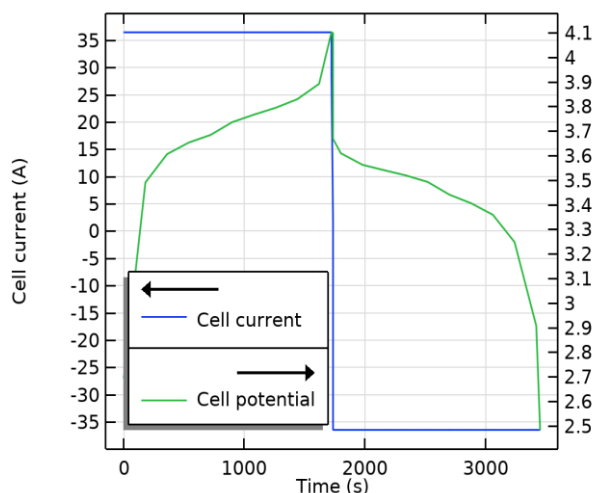
**Fig.5: load cycle at 60 $\mu\text{m}$  thickness**



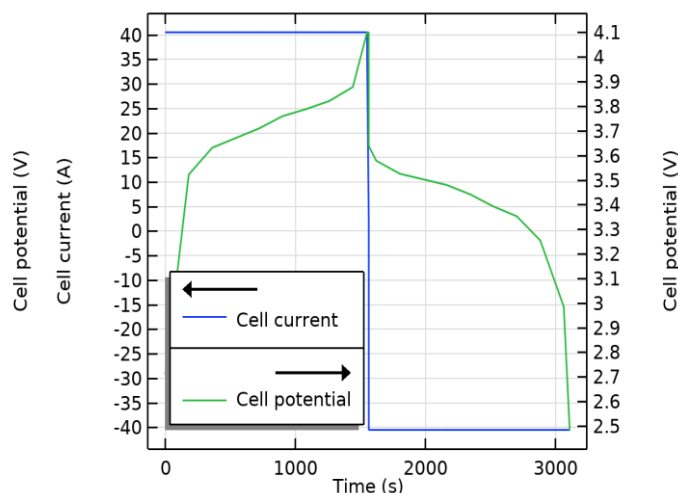
**Fig.6: load cycle at 70 $\mu\text{m}$  thickness**



**Fig.7: load cycle at 80 $\mu\text{m}$  thickness**



**Fig.8: load cycle at 90µm thickness**

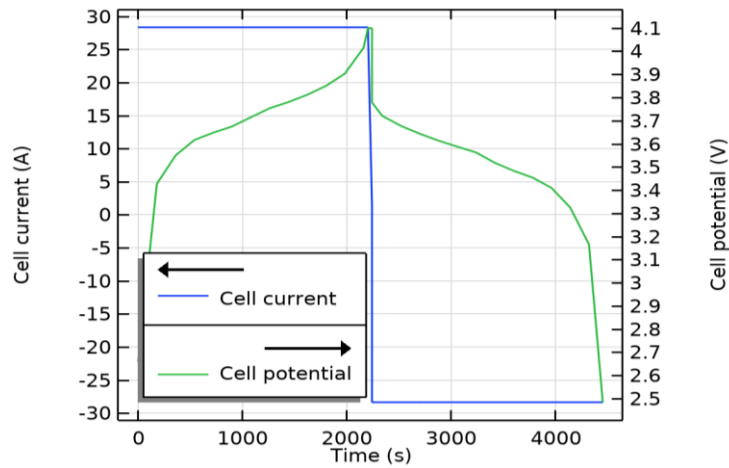


**Fig.9: load cycle at 100µm thickness**

In the figure above it is clearly visible that there is a hold of time between charging and discharging for the less thick NCA cathode. With the increasing thickness the hold is being reduced. But with the increasing thickness, the voltage drop is also increasing. Apart from these two factors, the charging time of the battery is decreasing with the increment of thickness. From fig.4 we can see that for thickness of 50µm the charging time is more than 3000 seconds, the same for the thickness of 80µm is around 2000s which is shown in fig.7, in fig.8 that is of NCA of thickness 100µm, the charging time is around 1500 seconds. So, it's clear that with increasing thickness the charging time is decreasing.

One more outcome is visible from the above figures, and it is that the voltage is pretty much constant for more time period in the battery with thicker cathode. As we can see in the fig.4, fig.5, fig.6 that the voltage is dropping linearly, whereas in fig.7, fig.8, fig.9 we observe that there is plateau like formation of the curve between voltage and time. So with increasing thickness, voltage vs time curve shows the plateau like formation.

The hold between charging and discharging is decreasing with the increase in thickness of the cathode, but voltage drop is high in the thicker cathode. The main reason found for this is the passivation layer and polarization of the lithium ions. Passivation layer is formed at the ends of the electrodes which prevents the conduction of the ions. With increasing thickness, the number of lithium ions produced increases which easily penetrates the passivation layer, but when these ions are less in number, it is difficult for them to penetrate quickly, which leads to the hold of time between charging and discharging. But the problem with the higher thickness is that, more the ions produced more the polarization effect. Yes, at the higher thickness we have seen voltage drop is more as compared to the lower thickness NCA cathode. [21,22] This voltage drop can be easily observed and compared from the fig.4 and fig.9. So, with increasing thickness hold between charge and discharge can be overcome but the polarization leads to voltage drop. So the optimized thickness as per this results is around 80µm [23]. As we can see in fig.7 that there is no hold between charging and discharging, and also there is an acceptable voltage drop. The voltage drop due to polarization at different thickness of cathode is given in table 3.



**Fig.10: load cycle at 75µm thickness**

For more precision we have plotted the charge and discharge curve of the battery with the cathode thickness of 75µm. And as per our observation it is similar to the curve, when the thickness of cathode was 80µm. The only major difference observed is the charging time. When the thickness of cathode was 80µm the charging time was less than 2000 seconds, but in case of 75µm thickness the charging time is more than 2000 seconds. All these can be seen clearly in fig.10

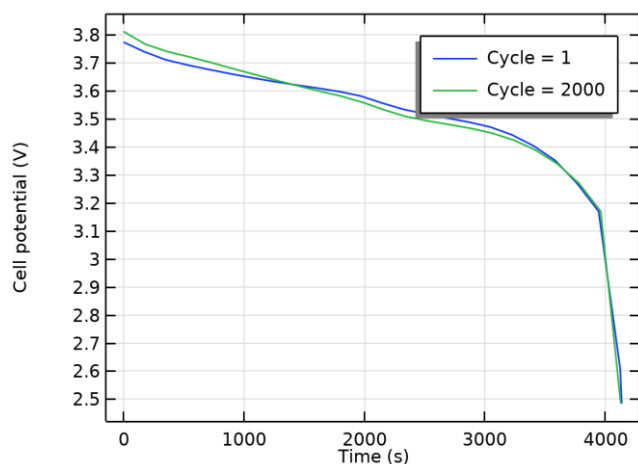
**Table 3: Voltage drop due to polarization at different thickness**

Thickness of cathode(µm)	Peak Voltage (V)	Nominal Voltage (V)	Voltage Drop $\Delta V$
50	4.1	4.0	0.1
60	4.1	3.87	0.23
70	4.1	3.78	0.32
75	4.1	3.78	0.32
80	4.1	3.71	0.39
90	4.1	3.68	0.42
100	4.1	3.64	0.46

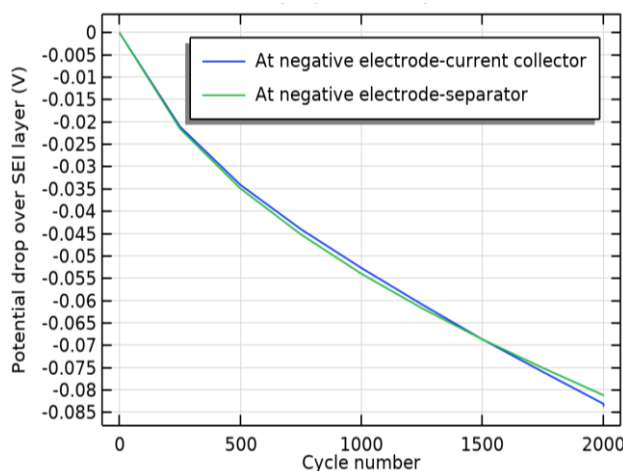
For further studies, we chose the thickness of 75µm and 80µm and studied the discharge curve at different cycle rates and voltage drops due to SEI layer formation at different cycle rates. The cycle rate at which the model was executed is, 0.5C, 1C, and 1.5C.



**At thickness 75 $\mu$ m and C rate 0.5C**



**Fig.11: discharge curve**

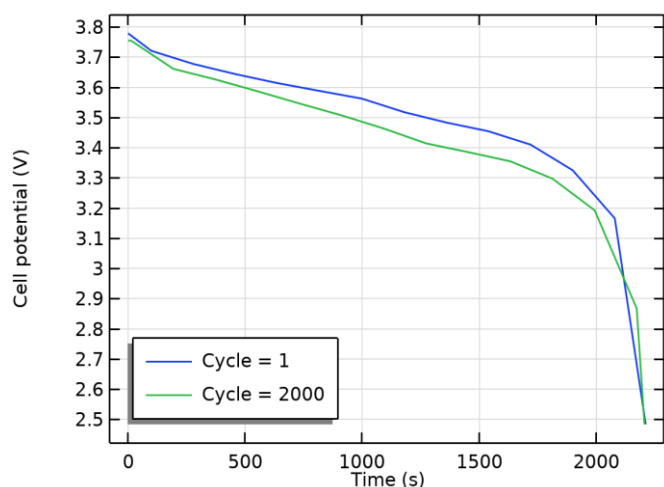


**Fig.12: potential drop due to SEI layer**

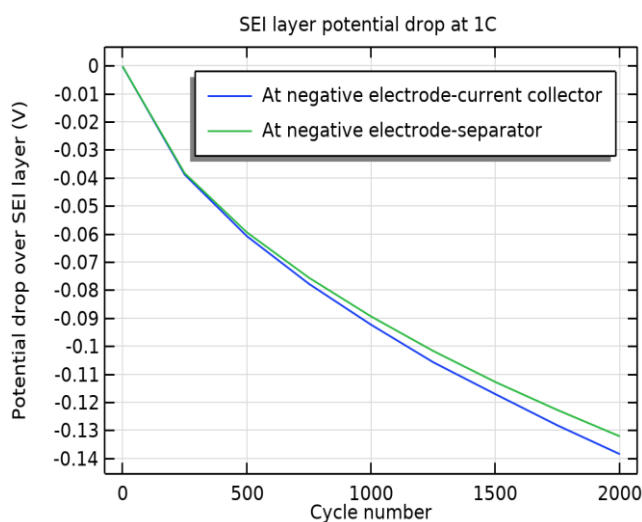
The fig.11 and fig.12 represents the discharge curve of the battery in 1<sup>st</sup> and 2000<sup>th</sup> cycle, and potential drop due to SEI layer formation respectively, with thickness of the NCA cathode 75 $\mu$ m and operated at 0.5C cycle rate. In fig.11 we can clearly see that the peak voltage in 1<sup>st</sup> and 2000<sup>th</sup> cycle is almost the same which is around 3.8V, there has been no drop in voltage with increasing cycle.

In fig.12 the two curves show the voltage drop due to SEI layer at anode collector end, and anode separator end. It is clearly visible that there is a significant voltage drop, with increasing cycles and at both ends it is almost equal.

**At thickness 75 $\mu$ m and C rate 1C**



**Fig.13: discharge curve**

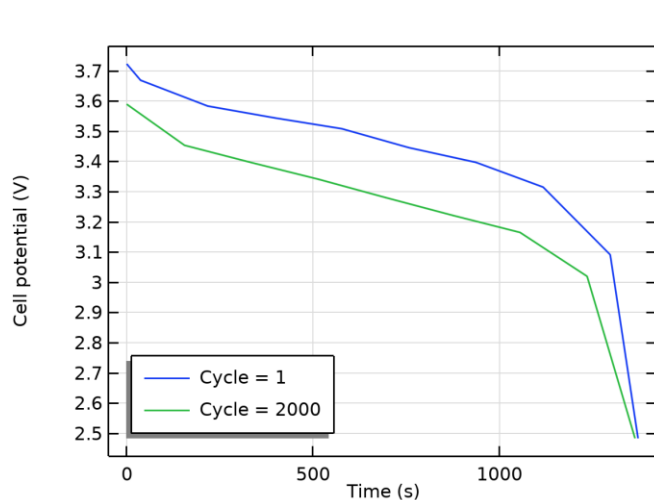


**Fig.14: potential drop due to SEI layer**

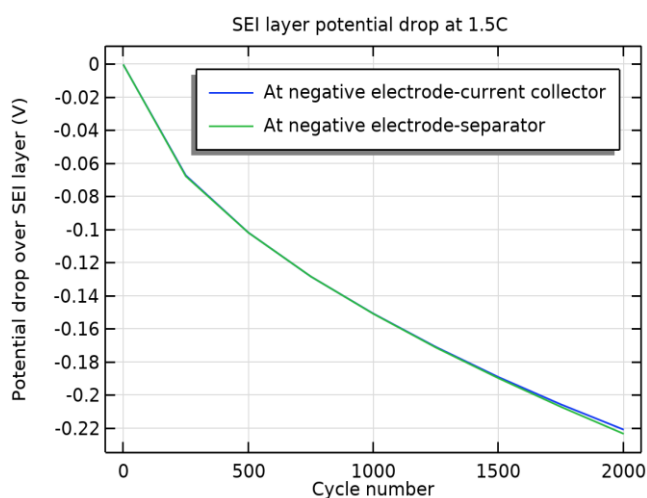
The fig.13 and fig.14 represents the discharge curve of the battery in 1<sup>st</sup> and 200<sup>th</sup> cycle, and potential drop due to SEI layer formation respectively, with thickness of the NCA cathode 75 $\mu$ m and operated at 1C cycle rate. In fig.13 we can clearly see that the peak voltage in 1<sup>st</sup> cycle is around 3.7V, whereas in 2000<sup>th</sup> cycle there has been a drop in voltage with time, the drop is about 0.1V

In fig.14 the two curves shows the voltage drop due to SEI layer at anode collector end, and anode separator end. It is clearly visible that there is a significant voltage drop, with increasing cycles the voltage drop at collector is more than that at separator end when battery is operated at 1C cycle rate. The change is observable after 750 cycles.

#### At thickness 75 $\mu$ m and C rate 1.5C



**Fig.15: discharge curve**



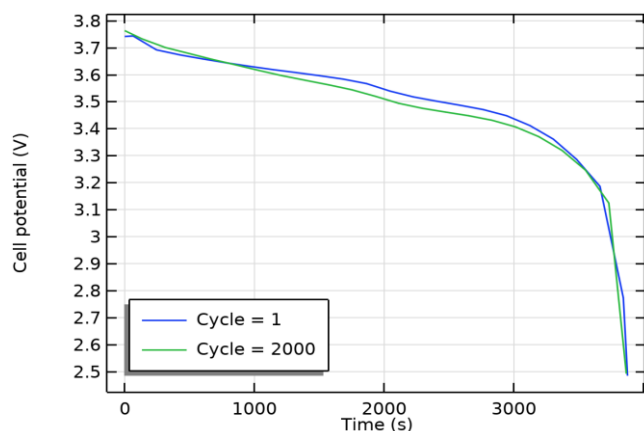
**Fig.16: potential drop due to SEI layer**

The fig.15 and fig.16 represents the discharge curve of the battery in 1<sup>st</sup> and 2000<sup>th</sup> cycle, and potential drop due to SEI layer formation respectively, with thickness of the NCA cathode 75 $\mu$ m and operated at 1.5C cycle rate. In fig.15 we can clearly see that the peak voltage in 1<sup>st</sup> cycle is 3.7V whereas in 2000<sup>th</sup> cycle the peak voltage is 3.6V, there has been a drop in voltage in 2000<sup>th</sup> cycle with passage of time, and it is about 0.2V.

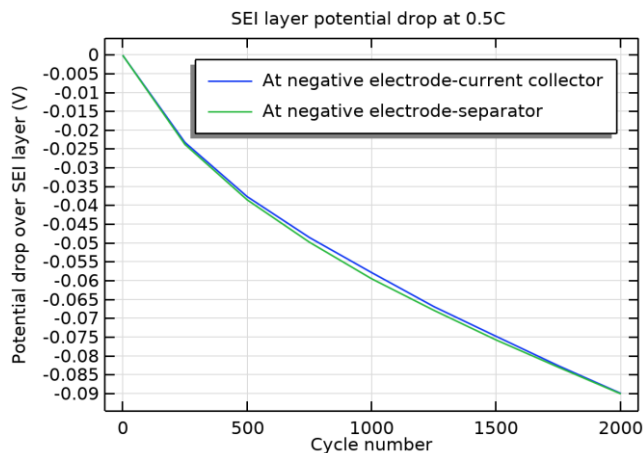
In fig.16 the two curves shows the voltage drop due to SEI layer at anode collector end, and anode separator end. It is clearly visible that there is a significant voltage drop, with increasing cycles and at both the ends it is almost equal, when operated at 1.5C cycle rate.

*After this, the discharge curve and voltage drop due to SEI layer formation were studied at the thickness of 80 $\mu$ m and with the cycle rate of 0.5C, 1C, and 1.5C, and their result is being explained further.*

**At thickness 80 $\mu$ m and C rate 0.5C**



**Fig.17: discharge curve**

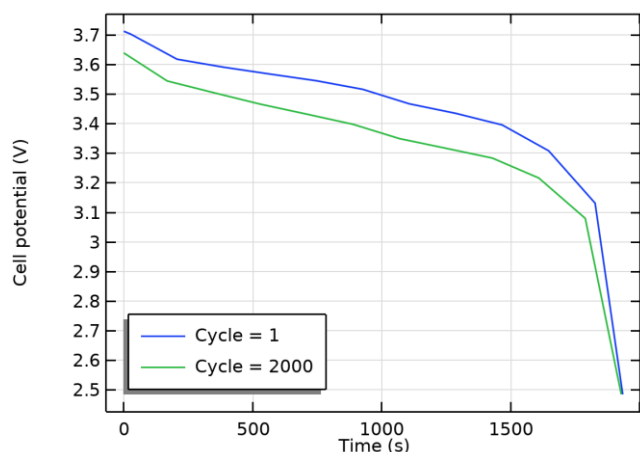


**Fig.18: potential drop due to SEI layer**

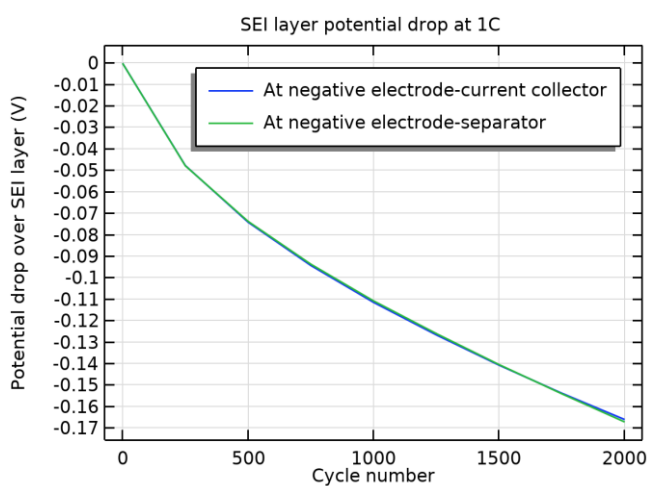
The fig.17 and fig.18 represents the discharge curve of the battery in 1<sup>st</sup> and 2000<sup>th</sup> cycle, and potential drop due to SEI layer formation respectively, with thickness of the NCA cathode 80 $\mu$ m and operated at 0.5C cycle rate. In fig.17 we can clearly see that the peak volatge in 1<sup>st</sup> and 2000<sup>th</sup> cycle is almost same which is above 3.7V, there has been very little or no drop in volatge with increasing cycle, the discharge curve is similar.

In fig.18 the two curves shows the voltage drop due to SEI layer at anode collector end, and anode separator end. It is clearly visible that there is a significant volatge drop, with increasing cycles and at both the ends it is almost equal, at both the ends.

**At thickness 80 $\mu$ m and C rate 1C**



**Fig.19: discharge curve**

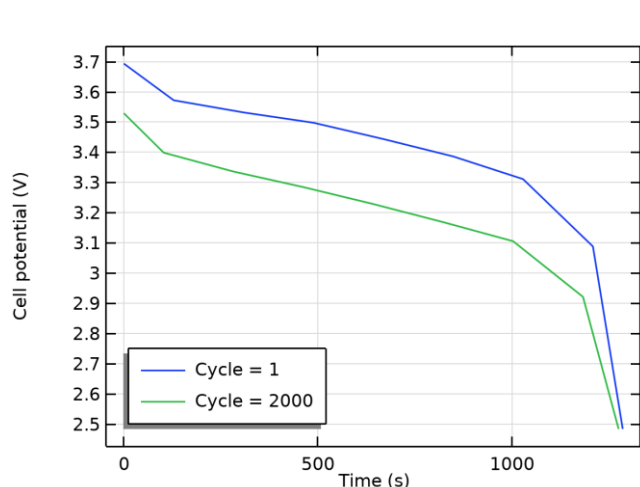


**Fig.20: potential drop due to SEI layer**

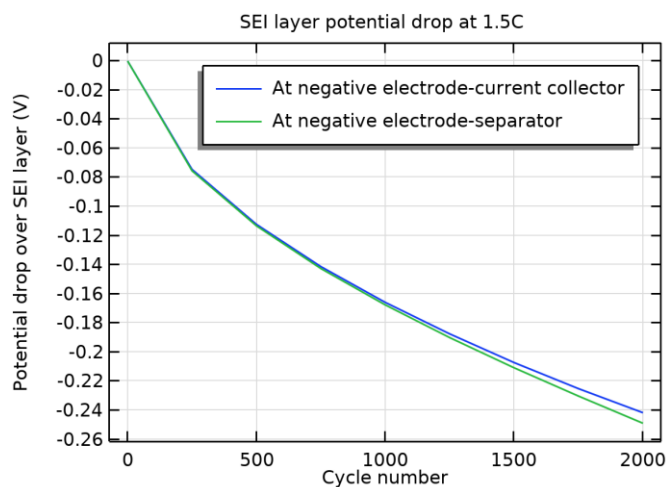
The fig.19 and fig.20 represents the discharge curve of the battery in 1<sup>st</sup> and 2000<sup>th</sup> cycle, and potential drop due to SEI layer formation respectively, with thickness of the NCA cathode 80 $\mu$ m and operated at 1C cycle rate. In fig.19 we can clearly see that the peak voltage in 1<sup>st</sup> cycle is 3.7V whereas in 2000<sup>th</sup> cycle the peak voltage is 3.6V. This 0.1V voltage difference slightly increases with the time as compared to 1<sup>st</sup> and 2000<sup>th</sup> cycle.

In fig.20 the two curves shows the voltage drop due to SEI layer at anode collector end, and anode separator end. It is clearly visible that there is a significant volatge drop, with increasing cycles and at both the ends it is almost equal, at both the ends.

**At thickness 80 $\mu$ m and C rate 1.5C**



**Fig.21: discharge curve**



**Fig.22: potential drop due to SEI layer**

The fig.21 and fig.22 represents the discharge curve of the battery in 1<sup>st</sup> and 2000<sup>th</sup> cycle, and potential drop due to SEI layer formation respectively, with thickness of the NCA cathode 80 $\mu$ m and operated at 1.5C cycle rate. In fig.21 we can clearly see that the peak volatge in 1<sup>st</sup> cycle is 3.7V whereas in 2000<sup>th</sup> cycle the peak voltage is 3.5V. This 0.2V voltage difference slightly increases with the time as compared to 1<sup>st</sup> and 2000<sup>th</sup> cycle.

In fig.22 the two curves shows the voltage drop due to SEI layer at anode collector end, and anode separator end. It is clearly visible that there is a significant voltage drop, with increasing cycles and at both the ends it is almost equal till 1250 cycles, then after the voltage drop at separator end increases by some fraction.

*Table 4: Capacity at different cycle rate and nominal voltage at first and 2000<sup>th</sup> cycle*

Thickness	C rate	Discharge Capacity (mAh/g)	Voltage in 1 <sup>st</sup> Cycle	Voltage in 2000 <sup>th</sup> Cycle
75 $\mu\text{m}$	0.5	180.30	3.81	3.78
	1	193.40	3.78	3.76
	1.5	180.03	3.72	3.59
80 $\mu\text{m}$	0.5	191.34	3.75	3.75
	1	171.17	3.70	3.64
	1.5	176.61	3.70	3.52

Capacity at different cycle rates with different thickness has been calculated and shown in the Table4, also the nominal voltage in first cycle and 2000<sup>th</sup> cycle at those cycle rates and thicknesses of the cathode has been presented in this table. The cathode with thickness 75 $\mu\text{m}$  has maximum capacity at 1C cycle rate, and also has very less drop in potential between 1<sup>st</sup> and 2000<sup>th</sup> cycle. The same result is observed in the cathode of 80 $\mu\text{m}$  thickness, but at 0.5C cycle rate.

## Chapter-6

### Conclusion

The project work covers the need of development of renewable source of energy, types of batteries, compared the different types of secondary batteries, advancement of Lithium-ion technology and its importance in the field of energy storage. The thesis extensively describes the strategies for selection of suitable thickness of NCA cathode and also have the reasons to choose NCA as a cathode material.

The theoretical simulation and modelling of 1D lithium-ion battery has been performed with variable thickness of cathode. Thickness of anode and separator was kept constant, and thickness of cathode was varied from 50 $\mu\text{m}$  to 100 $\mu\text{m}$  with increment of 10 $\mu\text{m}$ . The charge and discharge curve has been plotted for these thicknesses and it was observed that the battery with NCA cathode of thickness 75 $\mu\text{m}$  and 80 $\mu\text{m}$  shows the best results, as it has higher discharge time, discharge capacity and relatively low polarization effect. Further, the simulation was carried out at three different cycle rates, 0.5C, 1C, and 1.5C for these optimized thickness of 75 $\mu\text{m}$  and 80 $\mu\text{m}$ . Thereafter, the discharge curve was plotted for 1<sup>st</sup> and 2000<sup>th</sup> cycles at the above cycle rates. It was found that the battery with NCA cathode of thickness 75 $\mu\text{m}$  shows best results with 1C cycle rate and NCA cathode of thickness 80 $\mu\text{m}$  shows best result with 0.5C cycle rate. Moreover, the potential drop due to SEI layer is more prominent in battery with the thickness of NCA cathode of 80 $\mu\text{m}$ . Overall with all these observations it is concluded that the battery with NCA as cathode material performs best with the thickness of cathode about 75 $\mu\text{m}$  to 80 $\mu\text{m}$ .

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## Electrochemical performance of NCA based cathodes for variable thickness of electrode through modelling and simulation

Pritesh Singh, Vaishali Dudeja, Amrish K. Panwar\*

Department of Applied Physics, Delhi Technological University, Delhi 110042, India

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### ABSTRACT

In this study, modelling of lithium-ion battery (LIB) has been carried out using theoretical simulation. The simulation work has been performed for the better electrochemical performance of the battery with the  $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Al}_{0.1}\text{O}_2$  (NCA) type cathode material, Graphitic,  $\text{Li}_2\text{C}_6$  as an anode material, and  $\text{LiPF}_6$  as liquid electrolyte. Initially, the thickness variation of the cathode material of NCA has been performed. The cycle of the cells made of seven variable thicknesses of the NCA cathode has been estimated. Thereafter, the effect of optimized electrode thickness on the discharge curve of the cell at different C rates such as 0.5C, 1C, 1.5C has been obtained from the simulation results. The voltage drop at the cathode and anode ends due to the SEI layer has been simulated at the respective C rates for first ( $1^{\text{st}}$ ) and 2000<sup>th</sup> cycles. The results obtained from all three simulations show consistency with the previously reported literature based on experimental work. Hence, the cathode material (NCA) thickness is optimized and chosen easily according to the application and need.

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### 1. Introduction

Nowadays, the energies are generated by various non-renewable sources such as crude oil, coal, natural gases, etc. These sources pose a risk to the global atmosphere and climate change due to the production of  $\text{CO}_2$  gases, air pollution, and greenhouse gases [1]. To overcome these issues, the research work is going on in this area to produce renewable energy sources such as solar cells, photovoltaic cells, micro-wind turbines, water energy, etc. [2]. But due to the unavailability of regular source of energy from these. There is utmost requirement of good energy storage devices to provide the constant supply to the electric consumers, so that supply cannot be affected by the fluctuation of the renewable energy sources. Now a days rechargeable batteries are one of the most useful energy storage alternates to fulfil the constant supply demand.

Rechargeable battery is an electrochemical cell with a positive electrode as cathode and a negative electrode as anode, are separated by a solid separator and filled with liquid electrolyte and follow the multiple charge – discharge. Among various rechargeable

batteries, currently the lithium-ion battery (LIB) is most popular and useful technology in the battery sector. These batteries have high energy and power density, and better cycle capabilities than other types of rechargeable batteries. These qualities are very much important in the modern world and its applications like electrical and hybrid vehicles and one of the most important sectors i.e., energy storage systems which are used in renewable energy applications. The chemistry of the lithium-ion batteries follows the movement of lithium ions towards the positive end (cathode) through the organic electrolytic medium from the negative end (anode) during discharge process. Mostly, the anodes in lithium-ion batteries are made of graphite.

There are different types of cathode materials such as layered,  $\text{LiCoO}_2$ , spinel,  $\text{LiMn}_2\text{O}_4$  (LMO), olivine,  $\text{LiFePO}_4$  (LFP) and other compounds of similar structure [3,4,5]. Among the all types of developed cathode materials, the layered materials containing cobalt, nickel, and aluminum with different concentrations are of good capacity, good energy and power densities. As these different cathodes allow high lithium insertion and intercalation levels leading to large energy storage quantities. The lithium nickel cobalt aluminum oxides (NCAs) with variable composition of Ni, Co and Al content are a group of mixed metal oxides [6]. Some of them are important due to their application in lithium-ion batteries.

\* Corresponding author.

E-mail address: [amrish.phy@dtu.ac.in](mailto:amrish.phy@dtu.ac.in) (A.K. Panwar).

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NCA's are used as primary material on the positive electrode (cathode). The general formula is  $\text{LiNi}_x\text{Co}_y\text{Al}_z\text{O}_2$  with  $x + y + z = 1$ . The rechargeable lithium-ion batteries consisting of NCA cathodes are currently available in the market, and also being used in the electric vehicles (EVs) and electric appliances with  $x \approx 0.8$ . The batteries containing high Ni content in NCA cathode is referred as Ni-rich batteries. The voltage of those batteries lies between 3.8 V and 4.1 V, and has a nominal voltage of 3.6 V to 3.7 V. Hence, Ni-rich layered lithium transition-metal oxides,  $\text{LiNi}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$  (NCA) are archetypal cathodes used for EV batteries. Because of their high reversible capacity, good rate capability, and low cost. These materials have been studied extensively during the last two decades [7]. It has many applications such as in power grid, medical devices, and electric cars. Most notably Tesla, one of the leading manufacturers of electric vehicles, deploys NCA batteries in their electric vehicle's system. NCA batteries can store the same amount of energy as that of NMC batteries. Although NCA batteries are not widely used in the consumer industry, but are quite promising and used in the electric car industry.

The usable charge storage capacity of NCA is about 180 to 200 mAh/g, which is less than the theoretical storage capacity, 279 mAh/g [8]. However, the capacity of NCA is much higher than that of other alternative materials belonging to similar layered structure such as lithium cobalt oxide  $\text{LiCoO}_2$  (LCO) with 148 mAh/g, lithium iron phosphate  $\text{LiFePO}_4$  (LFP) with 165 mAh/g, and  $\text{LiNi}_{0.33}\text{Mn}_{0.33}\text{Co}_{0.33}\text{O}_2$  (NMC 333) with 170 mAh/g. Due to its high voltage, NCA enables batteries with high energy density. Another advantage of NCA is its excellent fast charging capability, High specific energy, good specific power, long life span. However, NCA materials also have demerits in terms of safety issues as there is need of extra safety features and circuitry.

Hence, the theoretical studies have been attempted on NCA based cathode material in lithium ion-battery. Therefore, a model has been prepared through the simulation with defined initial parameters. A one-dimensional (1-D) model is prepared with the variation of the thickness of the NCA cathode. Finally, the optimized thickness has been estimated to show the best electrochemical performance and to investigate polarization effect, charge-discharge voltage etc.

## 2. Model geometry (1-D) of battery

Fig. 1(a) indicates the 1-D model geometry used in the modelling of lithium-ion battery without electrolyte. Here, the length

starting from 0 to 5.5 represents the negative electrode i.e., Anode made of Graphitic Electrode,  $\text{Li}_x\text{C}_6$  material. After 5.5 to 8.5, the geometry shows the length of the separator and the last part from 8.5 to 15.5 is the length of the Positive electrode (Cathode) material used as  $\text{LiNi}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$  (NCA). Fig. 1(b) shows the whole cell geometry after dipped into the liquid Electrolyte,  $\text{LiPF}_6$  and it is shown by blue colour.

In this model, the thickness of the anode and separator is kept fixed as 55  $\mu\text{m}$  and 30  $\mu\text{m}$ , respectively [9]. Whereas the thickness of the cathode is varied. The initial parameters and the defined values used for the simulation as anode, separator, cathode and other parameters are given in Table 1.

## 3. Results and discussions

Fig. 2 shows the simulated data of charge and discharge of the battery at different thickness of NCA cathode. The Fig. 2(a), represents the charge-discharge curve of the battery for a thickness of the cathode, 50  $\mu\text{m}$ . The curve clearly shows that there is a hold between charging and discharging, and when the thickness is increased by 10  $\mu\text{m}$ , the hold reduces to some extent which can be seen in the Fig. 2(b) where the thickness of the cathode is 60  $\mu\text{m}$ . On further increment of the thickness by 10  $\mu\text{m}$  i.e., making the thickness of the cathode 70  $\mu\text{m}$ , the hold between charging and discharging has further decreased, but a few holding phenomena is still present when looked carefully depicted in Fig. 2(c). After taking the thickness to 80  $\mu\text{m}$ , 90  $\mu\text{m}$ , and 100  $\mu\text{m}$  the hold is completely reduced but there is a sharp fall in the discharging voltage, as we can see in Fig. 2(e), (f), (g), respectively. Fig. 2(d) represents the charge and discharge curve of the battery in which the thickness of cathode is 75  $\mu\text{m}$ , and it is clearly visible that the result is similar to that of the 80  $\mu\text{m}$  thickness curve but there was a slight hold between the charge and discharge curve. These curves also depict that with increasing thickness the voltage IR drop is decreasing and has a constant voltage for longer duration.

Not only there is variation in the hold, but with the increment of thickness, the time taken in the charging process is also being reduced. It is clearly visible that, for the thickness of 50  $\mu\text{m}$  the charging time is more than 3000 s and can be observed in Fig. 2 (a). Whereas, charging time for the battery having cathode of 80  $\mu\text{m}$  thickness is less than 2000 s, Fig. 2(e), and if we see about the cathode of 100  $\mu\text{m}$  thickness, the charging time is around 1500 s.

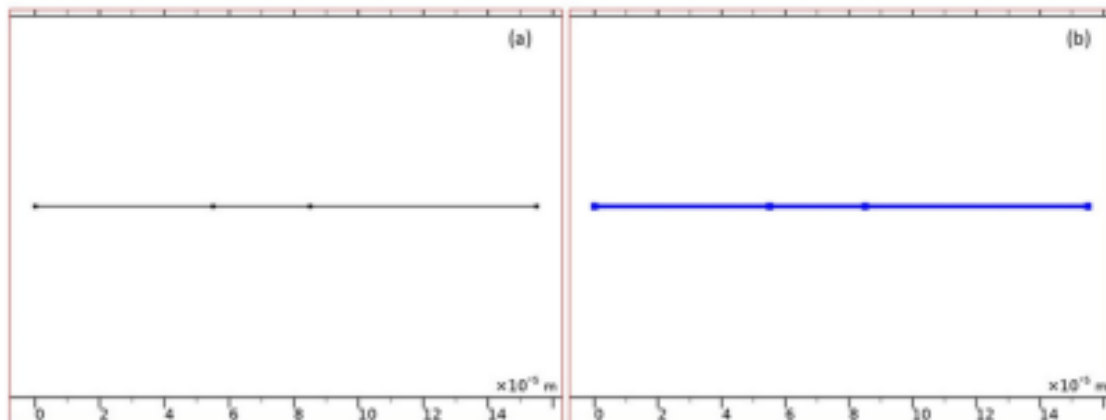


Fig. 1. (a) 1-D model geometry of anode, separator and cathode in battery, (b) 1-D model geometry of battery with electrolyte.

**Table 1**  
Initial Parameters of the model.

Description	Value	Anode	Cathode	Separator
Particle radius negative electrode	-	$2.5 \times 10^{-6}$ m	-	-
Particle radius positive electrode	-	-	$2.5 \times 10^{-7}$ m	-
Solid-phase volume fraction positive electrode	-	-	0.42	-
Electrolyte phase volume fraction positive electrode	-	-	0.41	-
Bruggeman coefficient for tortuosity in the positive electrode	-	-	2.98	-
Solid-phase volume fraction negative electrode	-	0.384	-	-
Electrolyte phase volume fraction negative electrode	-	0.444	-	-
Electrolyte phase volume fraction separator	-	-	-	0.37
Bruggeman coefficient for tortuosity in separator	-	-	-	3.15
Length of the negative electrode	-	$5.5 \times 10^{-3}$ m	-	-
Length of separator	-	-	-	$3 \times 10^{-3}$ m
Length of the positive electrode	-	-	$7 \times 10^{-3}$ m	-
Cell temperature	318.15 K	-	-	-
SEI layer conductivity	$5 \times 10^{-8}$ S/m	-	-	-
Initial SEI layer thickness	$1 \times 10^{-8}$ m	-	-	-
Number of cycles	2000	-	-	-

All these observations for variable thicknesses indicate that the thickness of about 80  $\mu\text{m}$  of the cathode material could be optimized for better discharge capacity and minimize holding step during discharge for this thickness. The theoretical results have been compared to experimental data, and it reveals that for better results, the thickness of the cathode was chosen between 70  $\mu\text{m}$  to 80  $\mu\text{m}$  [10]. If it is considered specially for the load cycle at 75  $\mu\text{m}$  thickness, apart from the hold between charging and the discharging, the charging time is more than 2000 s. Whereas for the thickness of 80  $\mu\text{m}$  the charging time is less than 2000 s.

There is a voltage hold for sometimes while discharging at low thickness whereas with increasing thickness the voltage hold is reduced but there is a drop in the voltage while increasing the thickness of NCA cathode. The main reason we have found for this is the fact that there is a passivation layer formed which leads to high impedance, and as at low thickness, the number of ions produced are also less as compared to the higher thickness materials [11,12]. The most of ions that are polarized may try to overcome the passivation layer, and its impedance leads to the hold of voltage, the rest of the polarized ions lead to voltage drop. Since there are a smaller number of ions, and most of them have been moved to overcome the passivation layer, the polarization is less that is why there is a low voltage drop. But with the increasing thickness, the number of ions produced increases which ultimately leads to heavy polarization, and this leads to more voltage drop. As there are more ions in thicker material, they overcome the passivation layer easily. The voltage drop due to polarization at different thickness of cathode is given in Table 2.

Thereafter, the discharge curve at different cycle rates and voltage drops due to SEI layer formation at different cycle rates for the thickness of 75  $\mu\text{m}$  and 80  $\mu\text{m}$  of cathode has been observed. Voltage in first cycle and 2000th cycle has been calculated at these two thicknesses with different cycle rate.

Fig. 3 shows the discharge curve and voltage drop due to SEI layer formation at different cycle rates, keeping the thickness of cathode material as 75  $\mu\text{m}$ . Fig. 3(a) shows the discharge curve of the battery at 1st cycle and 2000th cycle, battery is being operated at 0.5C rate. The peak voltage obtained was around 3.8 V in the 1st and 2000th cycle and also there has been no change in voltage with time, as almost similar discharge profile is observed. Fig. 3 (b) shows the potential drop at the negative electrode current collector and negative electrode separator end, due to SEI layer formation. With the increasing of cycle number, there has been a significant drop in the potential at both the ends and also the amount of drop is similar at both the ends. The discharge curve of the battery at 1st cycle and 2000th cycle when operated at a 1C rate is shown in Fig. 3(c). The peak voltage obtained was around

and slightly less than 3.8 V in the first and 2000th cycle but with time the voltage in the 2000th cycle is significantly less than the voltage in the first cycle, hence the difference is about 0.1 V. Fig. 3(d) indicates the potential drop at the negative electrode current collector and negative electrode separator end, due to SEI layer formation. With the increasing cycle number, there has been a significant drop in the potential at both ends, but after around 750 cycle numbers the potential drop at the negative electrode current collector end the potential drop is more than that at the negative electrode separator end, operated at 1C rate. The Fig. 3 (e) shows the discharge curve of the battery at 1st cycle and 2000th cycle when operated at a 1.5C rate. The peak voltage obtained was around 3.7 V in the first and 3.6 V in the 2000th cycle but with time, the voltage in the 2000th cycle is decreased by 0.2 V as compared to the first cycle. The Fig. 3(f) shows potential drop at the negative electrode current collector and negative electrode separator end, due to SEI layer formation. With the increasing cycle number, there has been a significant drop in the potential at both ends. The potential drop at both points is exactly equal with increasing cycle numbers. However, a slight change is visible after 1750 cycles.

Thereafter, the discharge curve and voltage drop due to SEI layer formation at different cycle rate at 80  $\mu\text{m}$  thickness of cathode is shown. Fig. 4(a) shows the discharge curve of the battery at 1st cycle and 2000th cycle, battery is being operated at 0.5C rate. The peak voltage obtained was around 3.7 V in the first and 2000th cycle and also there has been no change in voltage with time, i.e., in the first and 2000th cycle, the discharge curve is almost the same. The Fig. 4(b) shows the potential drop at the negative electrode current collector and negative electrode separator end, due to SEI layer formation. With the increasing cycle number, there has been a significant drop in the potential at both the ends and also the amount of drop is similar and somehow equal at both the ends. The Fig. 4(c) shows the discharge curve of the battery at 1st cycle and 2000th cycle when operated at a 1C rate. The peak voltage obtained was around and slightly less than 3.7 V in the first and 2000th cycle but with time the voltage in the 2000th cycle is significantly less than the voltage in the first cycle, the difference is about 0.1 V. The Fig. 4(d) shows the potential drop at the negative electrode current collector and negative electrode separator end, due to SEI layer formation. With the increasing cycle number, there has been a significant drop in the potential at both ends, but after around 750 cycle numbers the potential drop at the negative electrode current collector end, the potential drop is more than that at the negative electrode separator end, operated at 1C rate. The Fig. 4(e) shows the discharge curve of the battery at 1st cycle and 2000th cycle when operated at a 1.5C rate. The peak voltage

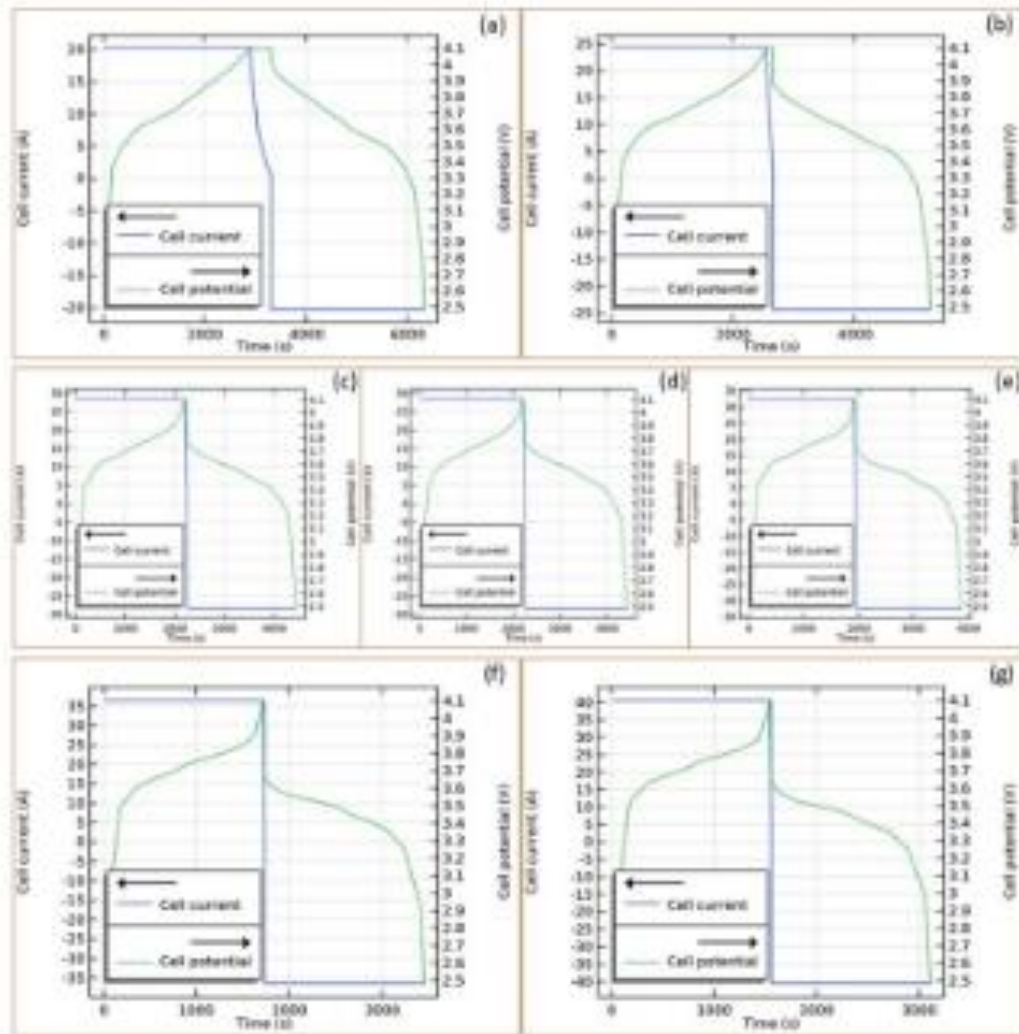


Fig. 2. Charge-Discharge curve of the battery at thicknesses of (a) 50  $\mu\text{m}$ , (b) 60  $\mu\text{m}$ , (c) 75  $\mu\text{m}$ , (d) 80  $\mu\text{m}$ , (e) 90  $\mu\text{m}$ , and (f) 100  $\mu\text{m}$  thickness of NCA cathode.

Table 3  
Voltage drop due to polarization at different thickness.

Thickness of cathode ( $\mu\text{m}$ )	Peak Voltage (V)	Nominal Voltage (V)	Voltage Drop (V)
50	4.1	4.0	0.1
60	4.1	3.87	0.23
70	4.1	3.78	0.32
75	4.1	3.78	0.32
80	4.1	3.71	0.39
90	4.1	3.68	0.42
100	4.1	3.64	0.46

obtained was around 3.7 V in the first and 3.5 V in the 2000th cycle but with time, the voltage in the 2000th cycle is decreased by 0.2 V as compared to the first cycle. The Fig. 4(f) shows potential drop at

the negative electrode current collector and negative electrode separator end, due to SEI layer formation. With the increasing cycle number, there has been a significant drop in the potential at both ends. The potential drop at both points is exactly equal with increasing cycle numbers. A slight change is visible after 1750 cycles. Voltage in first and 2000th cycles at different C rates has been calculated and given in Table 3.

#### 4. Conclusion

The theoretical simulation of NCA based cathode has been carried out for variable thickness of the material under 1-D cell geometry. The charge-discharge curve at variable thicknesses starting from 50  $\mu\text{m}$  to 100  $\mu\text{m}$  of the NCA based cathode material are obtained. It is estimated that the thicknesses 75  $\mu\text{m}$  and 80  $\mu\text{m}$

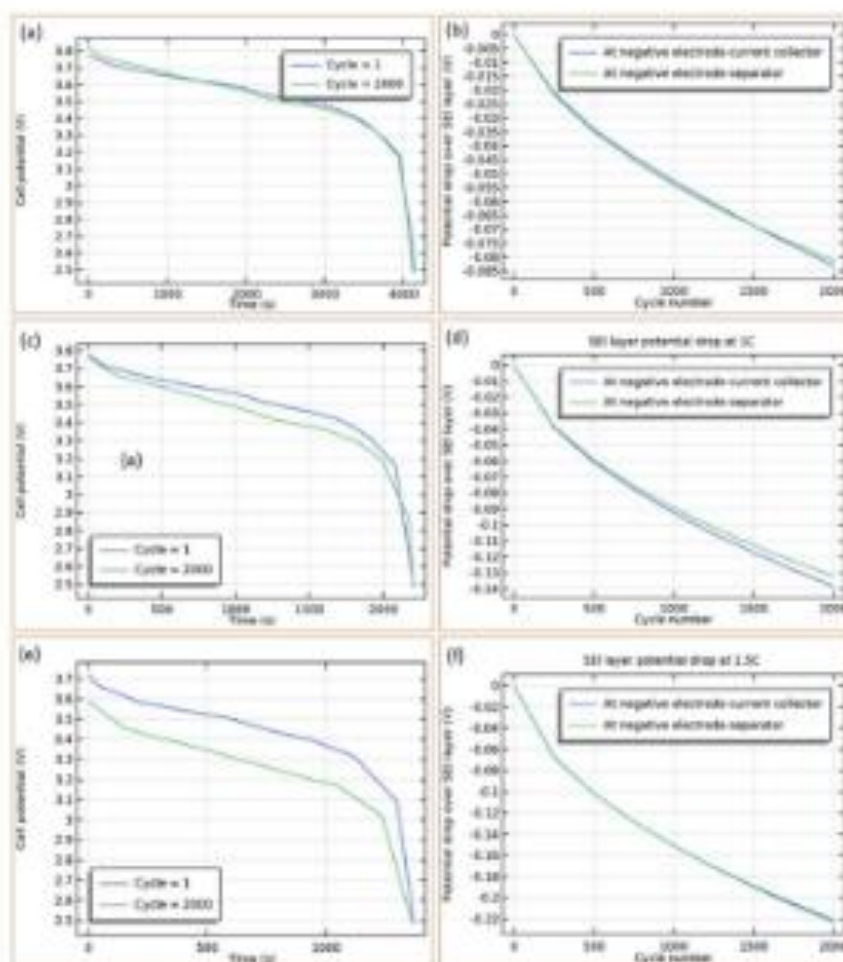


Fig. 3. Discharge curve and voltage drop due to SEI layer at (a, b) 0.5C rate, (c, d) 1C rate, and (e, f) 1.5C rate, for NCA cathode thickness of 75  $\mu\text{m}$ .

of NCA are found to be best for higher discharge time/ discharge capacity and relatively lower polarization effect comparing with higher thicknesses than 80  $\mu\text{m}$ . Further, simulated results were obtained for three different cycle rates: 0.5C, 1C, and 1.5C for these two thicknesses of 75  $\mu\text{m}$  and 80  $\mu\text{m}$ . Therefore, the discharge curve was obtained for 1st and 2000th cycles for both the thicknesses. Hence, it is observed that the polarization is increased with cycling up to 2000th cycle and it increases with increasing the discharge rates for both the thicknesses of 75  $\mu\text{m}$  and 80  $\mu\text{m}$ . Moreover, the effect on potential drop is by and large similar due to SEI layer and it is more prominent to 80  $\mu\text{m}$ . Overall with these observations, it is concluded that NCA cathode with a thickness between 70  $\mu\text{m}$  to 80  $\mu\text{m}$  performs highly well and may give better electrochemical performance.

#### ORCID iD authorship contribution statement

**Pritesh Singh:** Conceptualization, Methodology, Investigation, Data curation, Writing – original draft. **Vaibhavi Dedeja:** Conceptualization, Methodology, Investigation, Data curation, Writing – original draft. **Aashish K. Panwar:** Supervision, Formal analysis, Visualization, Writing – review & editing.

#### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

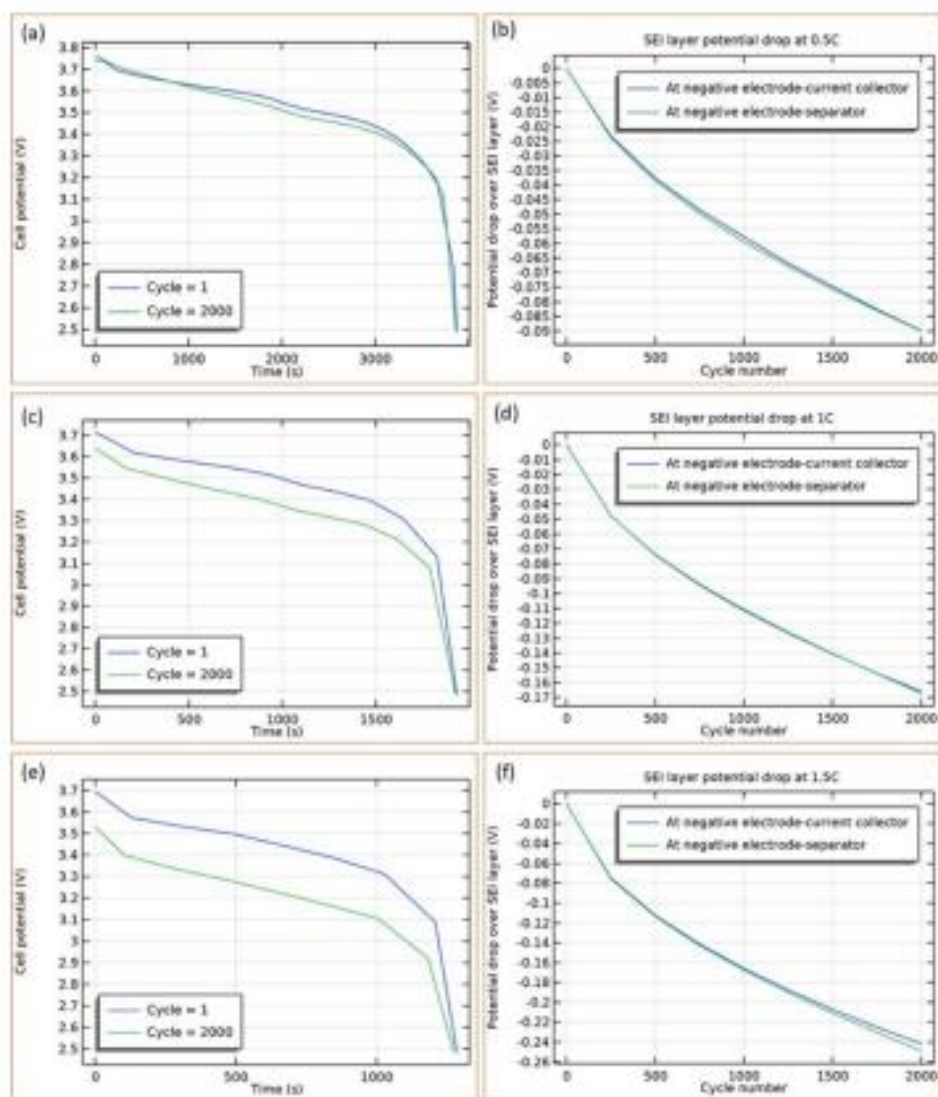


Fig. 4. Discharge curve and voltage drop due to SEI layer at (a, b) 0.5C rate, (c, d) 1C rate, and (e, f) 1.5C rate, for NCA cathode thickness of 80  $\mu\text{m}$ .

Table 3

Capacity at different cycle rate and nominal voltage at first and 2000th cycle.

Thickness	C rate	Discharge Capacity (mAh/g)	Voltage in 1st Cycle	Voltage in 2000th Cycle
75 $\mu\text{m}$	0.5	180.30	3.81	3.78
	1	193.40	3.78	3.76
	1.5	180.03	3.72	3.59
80 $\mu\text{m}$	0.5	191.34	3.75	3.75
	1	171.17	3.70	3.64
	1.5	176.61	3.70	3.52

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