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Comparative Study of NCM and NCA Electrode Material for Capacity-Fade Using 1-D Modeling

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ABSTRACT- Today, Lithium-ion (Li-ion) batteries are one of the most emerging power sources for almost all modern consumer electronic products. $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ (NCA) and $LiNi_{0.3}Co_{0.3}Mn_{0.3}O_2$ (NCM) are projected to be utilized in lithium-ion power batteries as two typical layered nickel-rich ternary cathode materials. Moreover, there is still a need for systematic study from an industrial aspect as to the advantages and drawbacks of these two nickel-rich materials. Hence, a comparative study of NCM and NCA electrode material for capacity-fade has been explored using a 1-D simulated model constructed in the multi-physics software. The capacity of a battery depends on the cell potential, discharge rate, state of charge (SoC), and state of health (SoH). Therefore, the comparison of these parameters and the cycle number of a battery is extremely important. During this comparative study of NCM and NCA electrode material, the capacity fade based on discharge rate, SoC, and SoH over cycle number of a battery has been reported.

Keywords: Lithium-ion battery, NCM, NCA, Capacity-fade, Battery Modelling.

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1. INTRODUCTION

Exploring energy storage and conversion device resources is critical to meet the growing demand for energy while also being environmentally friendly [1]. In recent years, the focus of rechargeable batteries has shifted to energy storage and conversion devices all around the world. Lithium-ion batteries (LIBs) are becoming more favored among rechargeable batteries because of their higher energy level and power densities when compared to other batteries. In addition to portable devices, these batteries are utilized in smart grids, the automobile industry, aeronautical engineering, communication, backup, and a range of other industries [2]. Battery usage is expanding on a massive scale across the globe for a variety of reasons, and battery dependency has been steadily increasing. Usually, Li-ion batteries undergo various electrochemical and mechanical changes during their working condition and are thus thought to be complex [1].

Electrochemical Energy Storage (EES) devices have become essential in contemporary times because of their high energy density, reliability, excellent life cycle, stability, etc. [3]. Lithium is the lightest metal with the most significant electrochemical potential, allowing for tremendous energy and power density. In LIBs, lithium metal is a promising anode material due to its small weight, high energy density, highest theoretical capacity, and low reduction potential. But uncontrolled reactivity with oxygen, moisture and interfacial interactions with the electrolyte limit the use of lithium metal an anode in LIBs for practical applications [2].

The application of these batteries is limited due to the physical limitations of the electrode materials whether it is for cathode or anode material. The electrodes in Li-ion batteries are made of various materials. Laptops and cell phones most commonly use the combination of Lithium cobalt oxide and graphite.

The battery industry is very much interested in finding ways to increase the energy, power, safety, life, and cost of the cathodes used in Li-ion batteries for automotive-propulsion applications. A novel strategy to build better batteries for hybrid electric, plug-in hybrid electric, and battery electric vehicles is to blend cathode materials. A 'blended' cathode's active material is a substantial composition of two or additional different lithium intercalation compounds. Blending of various compounds is motivated by creating a more balanced performance than is feasible with any single ingredient.

The ionic and electronic conduction properties of LIBs are portrayed by both the cathode and the anode electrode materials. It's difficult to improve the material's ionic conduction because it's so closely linked to the ionic transport species, which reveals the material's composition and structure. Specific technological procedures are utilized to boost the material's intrinsic electronic conductivity, such as adding a conduction additive or a carbon coating [3].

Among various developed cathode materials for lithium-ion batteries, NCM has a high energy density and capacity, which makes it an attractive candidate for usage in electric vehicles. The positive electrode's metal combinations offer stability to the cell while also making the electrode of high performance and cost-effective. Unlike other cathodes, NCM has a longer cycle life, better thermal stability, and a higher energy density [4,5]. In contrast to olivine structures with two phases, its layered cathode structure shows a single-phase intercalation process [6]. $Li(Ni_xMn_yCo_z)O_2$ is the cathode of NCM, with total molar fractions (x, y, and z) equaling one. [7]. In terms of similarity, the discharge chemistry of NCM is similar to $LiFePO_4$ (LFP) [8]. NCM is better for its low-temperature resistance [9]. Therefore, it may be better ordinate for low-temperature applications. However, lithium ion- batteries based on NCA cathode have aluminum instead of manganese in NCM cathode-based batteries. When compared to the NCM equivalent the results show that it improves the specific energy and lifespan of the previous [10].

Since, both the NCA and NCM have the same layered cathode structure and indicate many similar properties. When compared to the capacities of other structural cathodes $LiMn_2O_4$ (LMO) and LFP batteries, which are 148 $mAhg^{-1}$ and 170 $mAhg^{-1}$, respectively, the gravimetric capacity of NCA batteries is high with the value of 200 $mAhg^{-1}$. In general, the lifespan of NCA batteries are known to be long and lasting. NCA batteries are not considered to be safe when compared to other batteries and this becomes one of the major disadvantages for them. For usage in applications such as electric cars, these batteries require specific safety monitoring [11] NCA batteries have a similar discharge method or chemistry to other Li-ion batteries. For Lithium, oxidation occurs at the anode, while reduction occurs at the cathode, with charge reactions occurring from right to left and discharge reactions occurring from left to right. NCA allows for high energy density batteries due to the high voltage. Another advantage of NCA is its outstanding capacity to fast charging.

Both NCM and NCA have excessive applications in contemporary times. Their usage has been increasing and will show an upward trend exponentially in upcoming years. Thus, in this article, it is intended to compare the two in order to find the best-suited characteristics of each to further conclude best-suited applicability. The Multiphysics software used for the simulation of the two battery models is good to explore the cell potential versus time, that help in comparing the characteristics of NCM and NCA battery models.

In this investigation, a 1-D simulation has been performed to optimize the characteristics parameters of the battery using a computer model in multi-physics software. A theoretical model is constructed using experimental data to explore the lithium-ion battery's electrochemical, electrical, and thermal properties. The capacity variance with respect to a number of cycles, electrolyte potential, ion concentration on the surface, and electrolyte salt concentration are explored. Hence, a comparative study of NCM and NCA electrode material for capacity-fade has been explored using a 1-D simulated model.

2. BATTERY MODEL FORMULATION

Positive electrodes (PE) for lithium-ion coin cells have been studied using lithium nickel cobalt manganese oxide ($LiNi_{0.3}Co_{0.3}Mn_{0.3}O_2$) (NCM) and lithium nickel cobalt aluminum oxide ($LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$) (NCA). One-dimensional (1D) model geometry has been conducted for simulation analysis. The main focus of this simulation in both cases is on the coin cell's capacity retention, cell potential during various load cycles, and cell discharge pattern. These three components are monitored and serve as the model's control unit. This simulation model also depicts the charging and discharging curves and explains the cell's state of charge (SoC). Multiphysics software was used to create the lithium-ion coin cell model. A current collector (CC) on both sides, a separator, a positive electrode, and a negative electrode make up this model's simulation battery (NE) [12].

2.1 Model Parameters of NCM and NCA Electrodes

Initially, a 1D model of the battery here a coin cell, geometry was used and various frameworks were set based on the conditions of the battery. *Table 1* shows the several material criterion for studying the capacity of both NCA and NCM batteries, while *Table 2* shows the same for NCM batteries. Conditions for the boundary were arranged for the battery, one of them is thermal insulation in the leftmost boundary of the battery as well as in the center of the battery. As a result, the heat flux condition depicts the battery's heating surface, which is located on the rightmost boundary.

The salt concentrations, diffusion coefficient, and minimum and maximum state-of-charge of each electrode are entered as input parameters in the simulation software to study capacity retention for both materials.

1D battery model parameters to study capacity-related properties

Table 1: Material parameters for studying the capacity of NCA based batteries used in 1D battery simulation [13]

Property	Variable	Value	Unit
Diffusion coefficient	D_iso ; Dii = D_iso, Dij = 0	1.5e-15[m^2/s]	m^2/s
Electrical conductivity	sigma_iso ; sigmaii = sigma_iso, sigmaij = 0	91[S/m]	S/m
Reference concentration	cEeqref	48000[mol/m^3]	mol/m^3
Maximum electrode state-of-charge	Socmax	1	1
Minimum electrode state-of-charge	Socmin	0.25	1
Transport number	transpNum	1	1

Activity dependence	Fcl	1	1
Electrolyte conductivity	sigmal_iso ; sigmalii = sigmal_iso, sigmalij = 0	91[S/m]	S/m
Density	Rho	4740[kg/m^3]	kg/m^3

Table 2. Material parameters for studying the capacity of NCM based batteries used in 1D battery simulation

Property	Variable	Value	Unit
Diffusion coefficient	D_iso ; Dii = D_iso, Dij = 0	5e-13[m^2/s]	m^2/s
Electrical conductivity	sigma_iso ; sigmaii = sigma_iso, sigmaij = 0	100 [S/m]	S/m
Equilibrium potential	Eeq	Eeq_int1(SoC)+d EeqdT*(T-298[K])	V
Temperature derivative of equilibrium potential	dEeqdT	-10[J/mol/K]/F_const	V/K
Reference concentration	cEeqref	49000[mol/m^3]	mol/m^3
Max electrode SoC	SoHmax	0.975	1
Min electrode SoC	SoCmin	0	1
Electrolyte conductivity	sigmal_iso ; sigmalii = sigmal_iso, sigmalij = 0	9.65[S/m]	S/m
Transport number	transpNum	1	1
Activity dependence	Fcl	1	1

2.2 Model Parameters used for Negative Electrode and Electrolyte

Graphite is used as the anode material in the proposed simulated Lithium-ion cells in 1-D. Therefore, material characteristics parameters taken as input parameters for the NE (Graphite) are noted in *Table 3.*

Table 3: Material parameters used for Negative (graphite) electrode1-D battery simulation [12, 13]

Property	Variable	Value	Unit
Electrical conductivity	sigma_iso ; sigmaii = sigma_iso, sigmaij = 0	100[S/m]	S/m
Diffusion coefficient	D_iso ; Dii = D_iso, Dij = 0	1.4523e-13*exp (68025.7/8.314*(1/(T_ref/1[K])-1/(T2/1[K]))) [m^2/s]	m^2/s
Reference concentration	cEeqref	31507[mol/m^3]	mol/m^3
Max electrode SoC	SoCmax	0.98	1
Min electrode SoC	SoCmin	0	1
Electrolyte conductivity	sigmal_iso ; sigmalii = sigmal_iso, sigmalij = 0	468[S/m]	S/m
Transport number	transpNum	1	1
Activity dependence	Fcl	1	1
Thermal conductivity	k_iso ; kii = k_iso, kij = 0	1[W/(m*K)]	W/(m·K)
Heat capacity at constant pressure	Cp	750[J/(kg*K)]	J/(kg·K)
Density	Rho	2300[kg/m^3]	kg/m^3

The most common electrolyte used by manufacturers and researchers in Li-ion batteries to manufacture or research is LiPF$_6$. The electrochemical properties of cathode, and anode have been evaluated using the LIPF-EC-EMC electrolyte and works for an optimum composition for Li-ion batteries [14]. The details of electrolyte used in this study is given in *Table 4.*

Table 4: Input parameter used for electrolyte (LiPF6) [15]

Property	Variable	Value	Unit
Electrolyte salt concentration	cElsalt	1200[mol/m^3]	mol/m^3

3. RESULTS AND DISCUSSION

3.1 Related Capacity versus Cycle Number

The accurate estimation of the remaining battery life is critical for the system's reliable operation. As a result, SoH is introduced as a parameter. SoH is defined as a figure of standard for a battery's condition in comparison to its beginning conditions. SoH is measured in percent points, with 100 percent indicating the beginning state of a battery [16]. Cycle performance is used to determine the usable capacity of a battery. As the number of cycles increases, the usable capacity of the battery decreases [17].

In *Figure 1* for NCM, it can be noticed that for each cycle number the battery's usable capacity is changes. Based on cyclability, it can be observed that with an increase in cycle number the SoH decreases. At 400 cycles, cyclable lithium decreases from 1 to 0.98. It is observed that till 500 cycles, SoH drops more drastically than it decreases from 500 to 1800 cycles. For the next 400 cycles (400 to 800 cycles), It decreases mildly in comparison to the previous 400 cycles (0 to 400 cycles), and relative capacity decreases to 0.94. At 1800 cycles, it is decreased upto 0.91.

A C-rate is a measurement of how quickly a battery is depleted compared to its maximum capacity [18]. The discharge current at a 1C rate will totally deplete the battery in one hour. For the Nominal 1C rate the discharge capacity, it decreases throughout the cycles. It follows the same pattern as cyclable lithium. At 400 cycles, the Nominal 1C rate the discharge capacity decreases from 0.78 to 0.76. At 500 cycles, it decreases linearly with the cycle number. At 1800 cycles, it will decrease up to 0.67.

Figure 2 shows the change in usable capacity of a battery with number of cycles for NCA materials used as anode. Based on cyclable lithium, it can be observed that with increase in cycle number, SoH decreases. At 500 cycles, Cyclable lithium decreases from 1 to 0.95. It is observed that till 500 cycles, SoH drops more drastically than it decreases from 500 to 1800 cycles. For next 500 cycles (500 to 1000 cycles), it decreases mildly in comparison to previous 500 cycles (0 to 500 cycles) and relative capacity decreases to 0.92. At 1800 cycles, it will decrease up to 0.87.For Nominal 1C rate the discharge capacity, it decreases throughout the cycles. It follows the same pattern as cyclable lithium. Till 500 cycles, Nominal 1C discharge rate the capacity decreases from 0.88 to 0.84. At 500 cycles, it decreases linearly with the cycle number. At 1800 cycles, it will decrease up to 0.76.

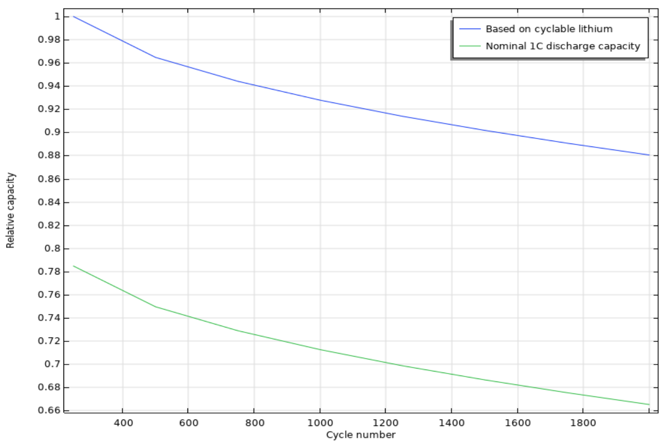

Figure 1: Variation of relative capacity with cycle number for NCM anode for SoH

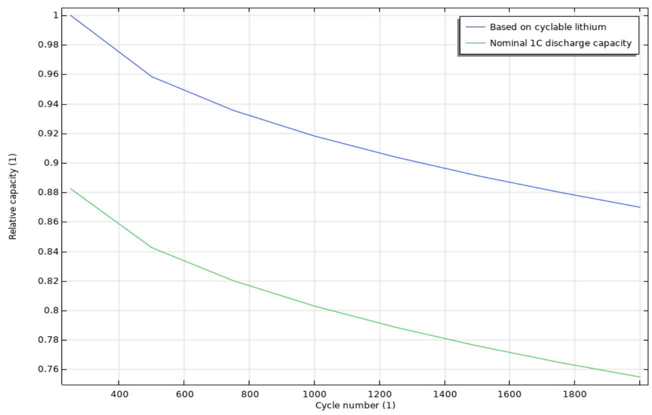

Figure 2: Variation of relative capacity with cycle number for NCA anode for SoH

3.2 Call Potential versus Time

To see the cyclic performance of the battery made of 1-D cathodes. The battery is run through 2000 cycles and the discharge curve is observed for both the NCM battery and the NCA battery model. The data of first and the last cycle is mentioned here. A linear drop has been observed for the first 500 seconds for both the cycles and the materials (NCM and NCA) [19]. In *Figure 3* for NCA, the decline follows a similar pattern till 2500 seconds after which a sudden drop in the cell potential is observed. For the first cycle, the steep decline is observed between 3.1V and 3.2V, whereas for the 2000th cycle, it is observed at 3.3V. Around 2700 seconds, the cell potential drops to a very low value for the 2000th cycle. In case of the 1st cycle, the same is observed post 3000s. A linear drop has been observed for the first 500 seconds for both the cycles as shown in *Figure 4* for NCM. The decline follows a similar pattern till 2000 seconds of the discharge (1C rate) after which a sudden drop in the cell potential is observed. For the first cycle, the steep decline is observed between 3.4V and 3.5V, whereas for the 2000th cycle, it is observed at 3.4V. Around 2200 seconds, the cell potential drops to a very low value for the 2000th cycle. In case of the 1st cycle, the same is observed post 2500s.

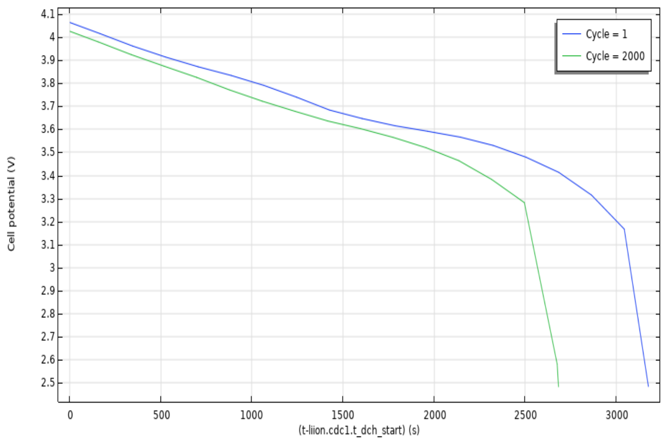

Figure 3: Discharge potential vs time for cycles 1st and 2000th for NCA

3.3 State of Charge versus Time

The SoC plays a crucial part in estimating the battery pack's remaining capacity. Because of the constrained battery model, accurate estimation of the SoC is quite complex and difficult to accomplish [20]. The battery's SoH is a crucial measure of its lifespan.

In *Figure 5* for NCM, the battery is run through 2000 cycles and the SoC curve is observed for both the NCM battery and the NCA battery model. The first and the last cycle is observed for the purpose of the study. The positive electrode in the battery for the first cycle starts from 0.73 whereas, for the last cycle, it starts from 0.63. At 3000 seconds, PE attains the lowest SoC for the first cycle as well as for the last cycle. After that, Positive electrode SoC at the first cycle remains closer to the last cycle with a minute difference.

For NE, SoC for the first cycle and last cycle is similar till the first 1000 seconds. After that, it can be observed that there is a minute difference between the first cycle and the last cycle. At 3000 seconds, both attain their highest SoC, for first cycle, it is 0.75 but for last cycle, it is closer to 0.6. Then there is a significant difference between first cycle SoC and last cycle SoC.

For NCA *Figure 6* shows that the battery is run through 2000 cycles and the SoC curve is observed for both the NCM battery and the NCA battery model. The positive electrode in the battery for the first cycle starts from 1 whereas for the last cycle, it starts from 0.88. At 3200 seconds, PE attain the lowest SoC for the first cycle as well as for the last cycle. After that, Positive electrode SoC at the first cycle remains closer to the last cycle with a minute difference.

For NE, SoC for the first cycle and last cycle is similar till the first 1000 seconds. Then it can be observed that there is a minute difference between the first cycle and the last cycle. At 3200 seconds, both attain their highest SoC, for first cycle it is 0.82 but for last cycle it is closer to 0.7. After that there is a significant difference between first cycle SoC and last cycle SoC.

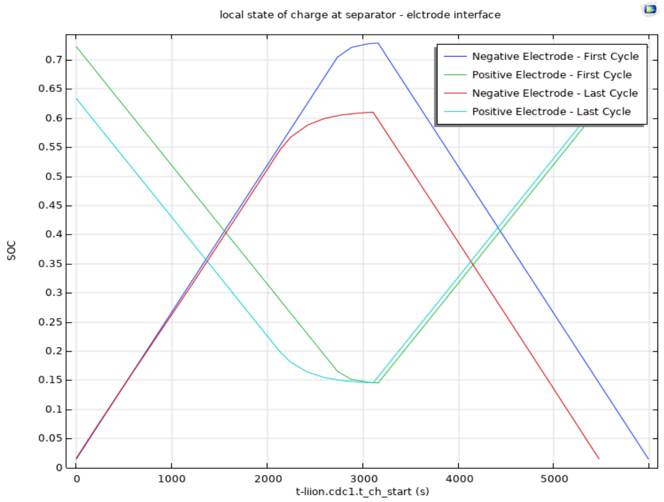

Figure 5: State of Charge vs Time for 5000s for NCM

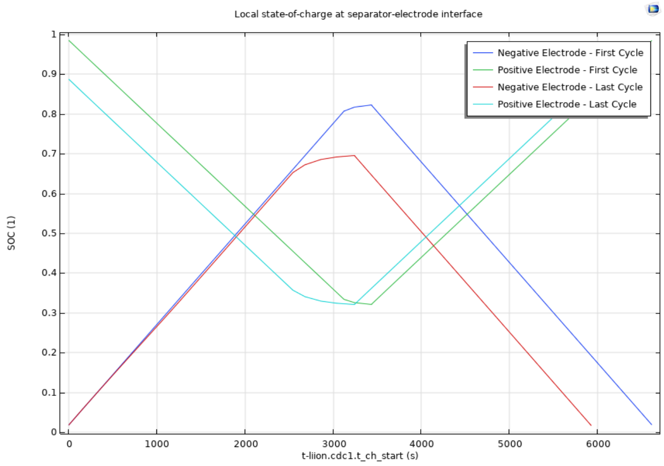

Figure 6: State of Charge vs Time for 5000s for NCA

4. CONCLUSION

This study reports the comparison of two typical layered nickel-rich ternary cathode materials (NCM and NCA electrode material) based on capacity fade of the battery. For capacity fade, the comparison is made in both the materials in terms of cell potential, discharge rate, SoC and SoH which affects the capacity of the battery. In the Relative capacity-cycle number curve, It has been observed that cyclable lithium capacity decreases by 1.23% (approx.) in NCA in comparison with NCM after completing 1800 cycles. While at Nominal 1C rate, discharge capacity decreases by 11.42% in NCM in comparison with NCA after completing 1800 cycles. In the SoC-time curve, the highest SoC for the NE (last cycle) reached 0.7 for NCA whereas between just crosses 0.6 for NCM, it indicates that higher charge retention in post multiple cycles for NCA is observed. Similarly, Cell potential shows a significant drop in the case of NCM for 2000 cycles as compared to NCA. Thus, the capacity fade models for the two positive electrode materials have been studied and summarized in a comparative manner.

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