32nd Electric Vehicle Symposium (EVS32)
Lyon, France, May 19 - 22, 2019

Fast charging method for Lithium ion batteries suppressing side reactions and lithium plating

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Summary

Reduction of charging time is one of challenging issues for different applications of lithium ion batteries. Increased charging current can reduce the charging time, but adversely accelerates degradation by enhanced side reactions and lithium deposition reaction. Currently suggested charging methods such as constant current and constant voltage (CC/CV) have not considered the fundamental mechanisms of ion transport and electrochemical reactions that degrade battery. Therefore, a reduced order electrochemical model considering degradation is developed for a large format NMC/Carbon lithium ion battery. The model is then used to estimate internal variables in real time that are used to determine the charging currents that consists of pulse and constant current and CV, which is experimentally validated using Battery-In-The-Loop (BIL). The results have shown that this proposed method enables not only reduction of charging time, but also suppression of side reaction and lithium plating and promotion of lithium stripping compared with the classical charging methods.

Keywords: lithium ion battery, fast charging, side reactions, lithium plating, lithium stripping

1 Introduction

Lithium ion battery is one of the most promising energy storage devices used for electric vehicles because of its high power and energy density. The long charging time presents one of technical barriers that should overcome for wide acceptance of electric vehicles in markets. Particularly, the reduction of charging time has brought to more attention as more battery is installed to extend the drive range. Super charger and extreme fast charger are the examples that provides a power up to 140kW and 350kW and the time to charge to 200 miles should take less than 36.55 min and 15.25 min, respectively.

In fact, the amount of charges that should be transported from cathode to anode electrode is an integral of the current applied over time. The charging time can be reduced by simply increasing the charging current, which however accelerates degradation of battery. Therefore, the challenging issues for designing a fast charging method are not only to reduce the charging time but also keep the degradation as low as possible.

The most widely used charging method is a combination of a constant current and constant voltage (CC/CV) charging method because of its simple and easy implementation to chargers [1]. However, the charging time cannot be significantly reduced by increasing the current of CC period because the cut-off voltage is reached earlier and correspondingly an extra time is required during the following CV charging. Additionally, a high charging C-rate leads to a high magnitude of the overpotential of the side reactions, which results in an increased side reaction rate and a significant reduction of cycle life [2]. The drawbacks of CC/CV charging method have been addressed and improved with respect to charging time, degradation, heat generation and by use of electric equivalent circuit models (EECM). These proposed charging methods can reduce the charging time, but do not consider the degradation effects. Physically, the degradation is directly related to internal variables of battery that include ion concentrations, potentials subject to operation conditions such as charging current and temperature. The currently proposed methods using empirical models or EECMs cannot provide the information on the internal variables. Therefore, we propose to use a reduced order electrochemical model (ROM) considering degradation effects in conjunction with advanced controls like Extended Kalman Filter (EKF) to estimate internal variables needed for limiting the actual rates of side reactions and lithium plating and promoting lithium stripping, whereby charging protocols are generated. In order to maximize lithium stripping, discharging pulse currents are used to recover ions out of the plated lithium.

According to investigations on degradation mechanisms of a large format pouch type lithium ion battery with NMC/Carbon or LFP/Carbon chemistries conducted by a research team at Auburn University [4], side reactions and lithium deposition reaction are the two main causes for degradation that take place at the surface of the anode graphite particles. The latter is also called lithium plating. Side reactions consume lithium ions and produce depositions on the anode surface, which leads to capacity loss and power loss. The lithium disposition reaction also takes place at the anode particle surface, which creates dendrites that potentially induces an internal short circuit and endangers the safety of battery system.

The lithium plating occurs when the local lithium deposition overpotential against a reference of Li/Li+ is less than 0V during charging, which creates a favourite condition. When the charging current becomes higher, lithium ions are saturated at the surface of anode particles because of relatively slow diffusion process in anode particles, which results in a high side reaction and lithium plating. Elevated temperature promotes side reactions, simply because of high mobility of ions and associated reactions. Consequently, capacity and power get faded [5]. The relationship between high charging current and lithium plating and side reaction is depicted in Figure 1

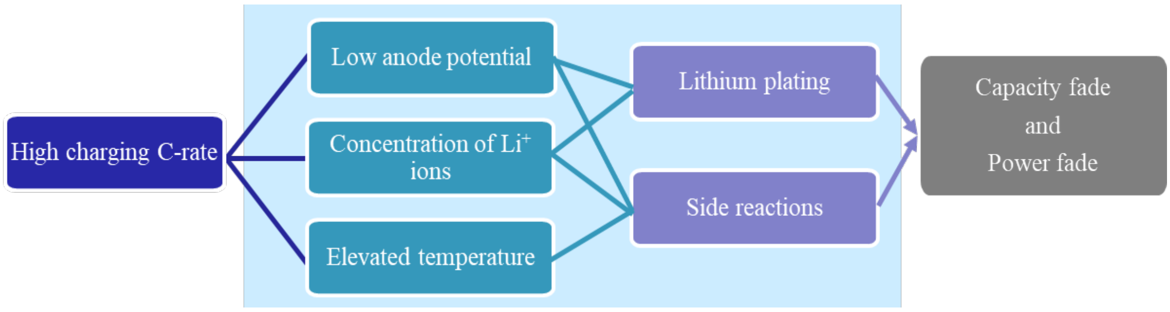


Figure 1: Relationship between high C rate and lithium plating and side reaction.

At the beginning of cycle life, degradation is dominantly caused by side reactions, where the capacity fades linearly. As the cycling continues, the side reactions deposit main products in the formed layer, which is called the solid electrolyte interphase (SEI) layer and located at the interface between particles and electrolytes. As a result, the SEI layer becomes growing and the porosity of anode electrode becomes less, which reduces anode ionic kinetics. Consequently, the gradient of the electrolyte potential increases and the anode potential becomes decreased, which enhances the lithium plating. The decrease of anode porosity and the increase of lithium plating overpotential due to the lithium plating amplify each other, so the aging process becomes accelerated and the capacity fade drops rapidly. These mechanisms lead to a transition of the capacity fade from linear to nonlinear range [6].

Hence, in order to limit both reactions, internal variables should be estimated which is only possible using electrochemical model, where the two reaction models are incorporated. The model is used to continuously estimate the anode overpotential and ion concentrations that are compared with limiting values to determine the charging currents, which delivers a protocol. At the same time, additional discharging pulse currents are applied to promote lithium stripping effects and recover ions out of the plated lithium, if formed.

2 Reduced Order Electrochemical Model (ROM) with EKF and Degradation model

2.1 ROM with EKF

When batteries are charged and discharged, ions are transported from cathode to anode electrode and vice versus, and intercalation and deintercalation take place. These processes can be mathematically described using a full order electrochemical model (FOM) that include mass transport, electrochemical kinetics, diffusions, overpotentials, and the associated potentials. The mass and charge transfer processes are governed by Fick's law and Ohm's law, respectively. The electrochemical kinetics are governed by the B-V equation under consideration of both anodic and cathodic reactions. The governing equations are composed of partial differential equations (PDE) and nonlinear equations that are coupled each other. Such equations can be solved only numerically, and the calculation time of the needed variables is too long to use for design of charging algorithm. Therefore, mathematical treatments of the equations are carried out, where the PDE is converted into ODE and the nonlinear equations are linearized, and very slowing parameters are assumed to be constant, which is called a reduced order model (ROM). The reduction of the order of the equations of ion concentration of electrode phase, ion concentration of electrolyte phase, electric potential of electrolyte phase and current density are performed by applying polynomial approach, state space approach, simplification and linearization, respectively as summarized in Figure 2. The detailed description of the model reduction approaches can be found in [7].

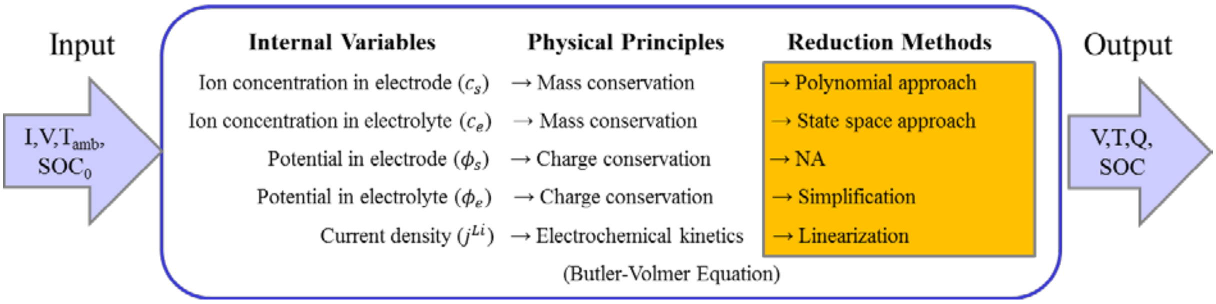


Figure 2: Schematic diagram of model principles.

Since the ROM includes measurement and state white noise and nonlinear functions, an Extended Kalman Filter (EKF) is employed to recursively optimize the noise and improve the dynamic error of average ion concentration and resulting SOC error given by the initial values [8].

2.2 Degradation models

Side reactions and lithium plating are the two major causes for degradation that are considered. In addition, lithium stripping is considered that dissolves the plated lithium metal and releases ions under certain operating conditions.

2.2.1 Side reaction

Side reaction is a reduction process between electrolyte solvent and lithium ions at the anode particle surface, which leads to capacity and power fade. The rate of side reactions, j_{side}^{Li}, is calculated using the B-V equation:

$$j_{side}^{Li} = -i_{0,side} a_s \exp\left(-\frac{\alpha_{c,side} n_{side} F}{RT}\eta_{side}\right) \qquad (1)$$

,where n_{side} is the number of ions involved in the side reactions that is equal to 2. η_{side} is the overpotential of side reactions.

The total amount of consumed ions is obtained by integration of reaction rate over the volume of the composite anode and time,

$$q_{ionloss}^{SR}(\tau) = \int_{x=0}^{\delta_-} \left(\int_{t=0}^{\tau} j_{side}^{Li}(x,t)dt \right) Adx \tag{2}$$

, $q_{ionloss}^{SR}$ is the loss of lithium ions with the unit of A·h and δ_- is the thickness of the composite anode and A is the cross section area of the battery.

Side reactions increase the ionic resistance of SEI layers and decrease the accessible surface area and porosity, which leads to the power fade [4]. Large amount of deposits are particularly produced at the interface between the composite anode and the separator to form a “deposit layer”, because the side reaction rate near the separator is larger than that inside of the electrode particles. The deposits can clog the pores of the particles that decrease the accessible surface of the active material for charges. If some of the particles are fully covered by the deposits, they will be electrically isolated from other particles and participation in chemical reaction is not possible. Additionally, the irreversible side reactions also consume electrolyte solvent, which results in the decrease of the electrolyte volume fraction and consequently the ion conductivity in the electrolyte [9].

2.2.2 Lithium plating and stripping

Lithium plating produces a metallic solid lithium by reaction between the lithium ions and electrons during charging. This reaction consumes lithium ions. When the local potential of anode electrode against a reference of Li/Li+ is less than 0V, which is the equilibrium potential for lithium plating [10][11], the lithium plating is likely to occur. Deposition of lithium metal covers the surface of particles in the anode electrode and decreases the active area of particles, which leads to capacity fade because of its impermeable characteristic to ions.

Lithium stripping is a dissolution reaction that takes place during discharging, where the plated lithium metal is dissolved. This reverse reaction allows ions to recover from the plated lithium. However, plated lithium metal covers the particles and quickly reacts with the solvents of electrolytes because of its highly reactive property, and becomes insoluble products that are called the secondary SEI. In addition, if SEI or secondary SEI fully cover lithium metal that cannot be dissolved by lithium stripping, “dead lithium” is produced.

The reaction rate of the lithium plating phenomena can be calculated using the B-V equation when the lithium plating overpotential is less than 0V. In addition, the amount of the ion loss by the lithium plating can be calculated from the reaction rate.

$$j_{LiP/S}^{Li} = a_{s,Li} i_{0,Li} \left[\exp\left(\frac{\alpha_{a,Li} F}{RT} \eta_{LiP/S} \right) - \exp\left(-\frac{\alpha_{c,Li} F}{RT} \eta_{LiP/S} \right) \right] \tag{3}$$

, where $i_{0,Li}$ is the exchange current density. $\alpha_{c,Li}$ and $\alpha_{a,Li}$ are set to be 0.3 and 0.7 [12] due to the secondary SEI generation and dead lithium.

The overpotential of the lithium plating is expressed by

$$\eta_{Li} = \varphi_s - \varphi_e - U_{eq,Li} - \frac{R_{SEI,total}}{a_{s,Li}} j_{total}^{Li} \tag{4}$$

$$\eta_{LiP} = \begin{cases} 0 & for\, \eta_{Li} \geq 0 \\ \eta_{Li} & for\, \eta_{Li} < 0 \end{cases} \qquad \text{for lithium plating}$$

, where $a_{s,Li}$ is the specific active area of the lithium plating.

The total number of ion loss consumed by the lithium plating and the secondary SEI is obtained by integration of the reaction rate over the composite anode during a given time,

$$q_{ionloss}^{LiP}\left(\tau\right)=\int_{x=0}^{\delta_-}\left\{\int_{t=0}^{\tau}\left(1-\lambda\right)j_{LiP}^{Li}\left(x,t\right)dt\right\}Adx \tag{5}$$

$$q_{ionloss}^{SEI,\sec}\left(\tau\right)=\int_{x=0}^{\delta_-}\left\{\int_{\tau=0}^{t}\lambda\, j_{LiP}^{Li}\left(x,\tau\right)d\tau\right\}Adx \tag{6}$$

, where λ is the ratio for the amount between the plated lithium and the secondary SEI layer formed from the plated lithium.

In this model, the generation ratio of the secondary SEI from the plated lithium is assumed to be constant. The consumption of active material and solvents of electrolyte from the lithium plating and secondary SEI generation can be calculated from the reaction rate, which leads to a decrease of the volume fraction of active material and electrolyte. In addition, the increase of thickness of deposit layer is calculated, which results in an increase of SEI resistance and deposit layer resistance.

A model for the lithium stripping is developed under the following assumptions;

- Lithium stripping takes place only when plated lithium is already generated.
- If the plated lithium is completely dissolved, no more lithium stripping exists.
- Once the plated lithium reacts with EC and generates the secondary SEI, lithium stripping cannot take place.
- The rate of the secondary SEI formation from lithium plating is constant.

Likewise, the overpotential for the lithium stripping is defined by

$$\eta_{LiS}=\begin{cases}\eta_{Li} & for\,\eta_{Li}\geq 0\\ 0 & for\,\eta_{Li}<0\end{cases} \qquad \text{for lithium stripping}$$

, where η_{Li} is calculated according to the equation (4). Because of the lithium stripping, the total amount of recovered lithium ions can be obtained by integration of reaction rate over the composite anode and time.

$$q_{ionrecover}^{LiS}\left(\tau\right)=\int_{x=0}^{\delta_-}\left\{\int_{\tau=0}^{t}j_{LiS}^{Li}\left(x,\tau\right)d\tau\right\}Adx \tag{7}$$

Finally, the total ion loss by the lithium deposition reaction is the sum of individual loss that is given by the equations (5), (6) and (7);

$$q_{ionloss}^{Li}\left(\tau\right)=q_{ionloss}^{LiP}\left(\tau\right)+q_{ionloss}^{SEI,\sec}\left(\tau\right)+q_{ionrecover}^{LiS}\left(\tau\right) \tag{8}$$

3 Design of fast charging method

Block diagram of the proposed fast charging method is depicted in Figure 3. The calculated terminal voltage by the ROM compares with the measured terminal voltage. The voltage errors are corrected by EKF. The two degradation models are incorporated into the ROM. The ROM-EKF with the degradation models are used to calculate the rates of side reaction, lithium plating and lithium stripping, which are again used to continuously update the slowly varying five parameters for the degradation models that include the change of the volume fraction of active materials, electrolyte, capacity and resistances of SEI and deposit layer. Estimated ion concentrations of solids and potentials are used to calculate SOC and the rates of side reaction and lithium deposition reactions. Once the reference value for required SOC is given, the charging algorithm generates a series of charging currents that take into account a set of different limitations such as cutoff voltage and current, maximum surface ion concentration, the rates of the side reaction and lithium plating. The charging current consists of pulse, multi-stage of currents and constant voltage.

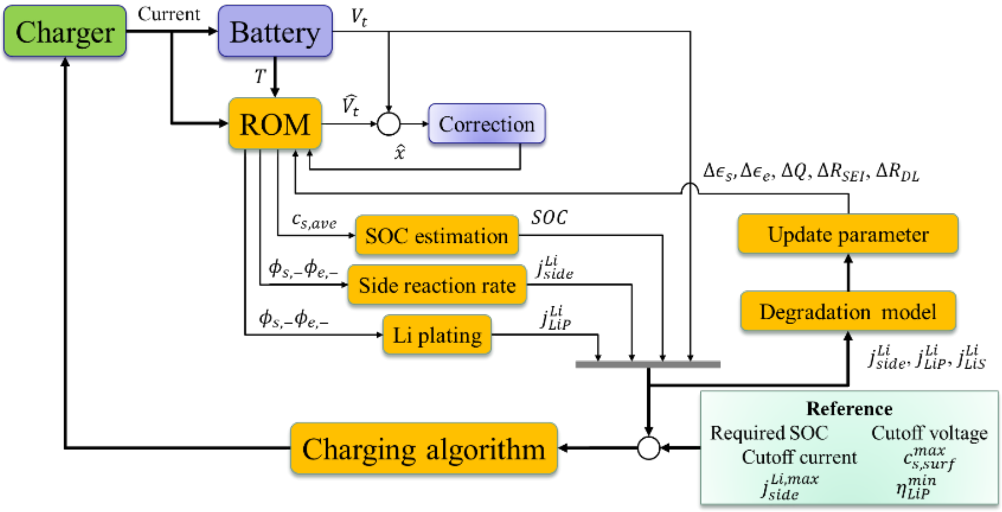


Figure 3: Block diagram for a fast and safe charging method.

A flowchart for the designed charging algorithm is depicted in Figure 4. Once the reference values for a requested SOC, cutoff voltage, maximum surface ion concentration, maximum side reaction rate, anode potential are given, a charging protocol is generated by comparing the values with those of the estimated and measured. At the beginning, a maximum C-rate is applied until one of the variables reaches its upper limitation. Upon reaching the limitation, the charging C-rate is reduced and kept as a constant, according to a predefined ΔSOC, which is repeated until the conditions of stop charging are fulfilled. The details of how to find the reference values are shown in [13]. In order to better control the surface ion concentration, extra resting periods are added to reduce the numbers of ions transported and gives the ions extra time to diffuse through the particles and to be intercalated [14]. The duration of the resting period is determined by considering the gradient of ion concentration in the composite anode.

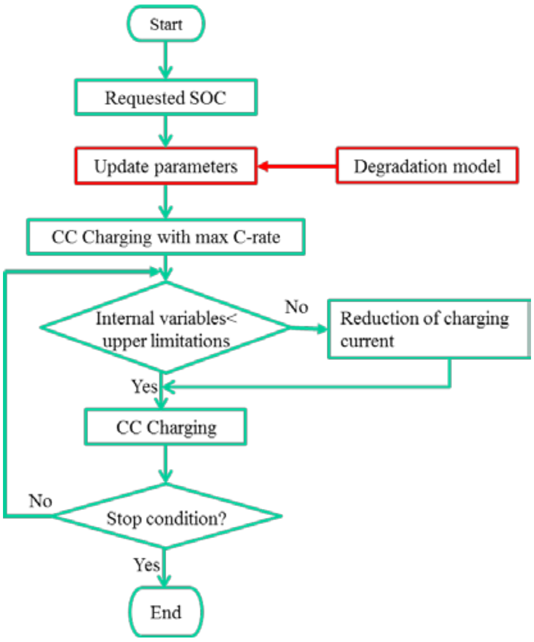
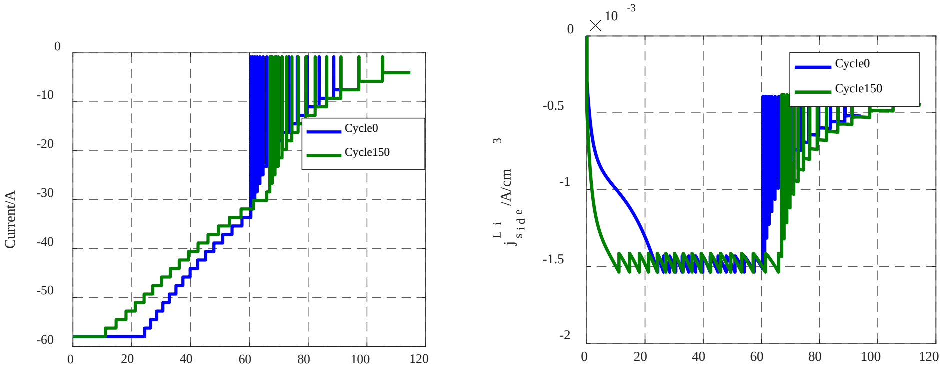


Figure 4: Flow chart of the proposing charging method.

3.1 Side reaction rate

The charging protocol under consideration of the maximum side reaction rate and the corresponding side reaction rate at BOL and 150 cycles are calculated and plotted in Figure 5. When high charging currents are applied, the side reation rate increases rapidly until the max allowed reaction rate and then the currents get decreased not to exceed the limitation. When battery gets degraded, the internal parameters such as resistances of SEI layer and deposit layer, active material volume fraction, and others change and the limitation is reached faster, which leads to an increase of the charging time. The set value is the same as that of the side reaction rate at 40% SOC of fresh cell. On the other hand, the surface ion concentration in high SOC range is almost kept as constant, so currents are pulsed to prevent any ovvershoot of the suface ion concentration.

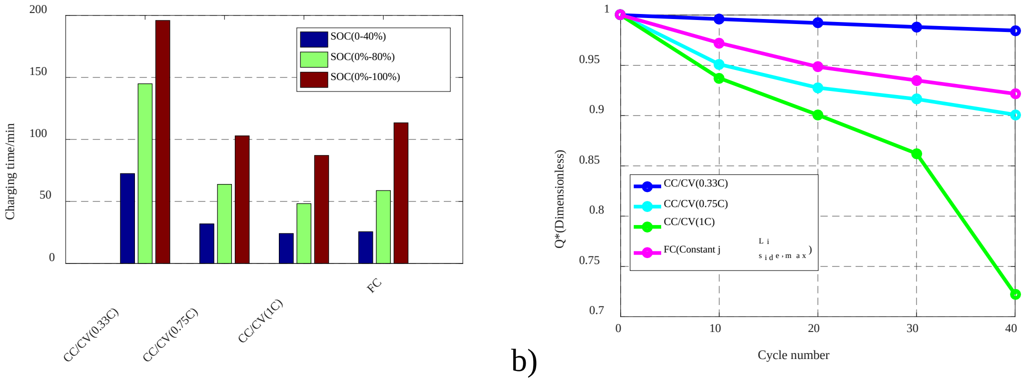


a) b)

Figure 5: Calculated charging protocol considering a constant maximum side reaction rate at cycle 0 and cycle 150. a) Current; b) side reaction rate.

The proposed charging method is implemented in the test station by integrating the ROM-EKF into LabVIEW using a MATLAB script. The ROM-EKF facilitates estimation of the internal variables like SOC, ion concentrations, and the side reaction rate based on the current and terminal voltage, which is used to constrain the charging current and generate the charging protocol for the requested SOC. In addition, the battery was placed in a tailor-made calorimeter that dynamically rejects the heat generated by the battery [15], which allows for minimization of the temperature effects on degradation. The maximum temperature variation becomes less than 0.3^{o}C even at a high heat generation rate.

The capacity of the cells is measured after every 10 cycles using the 1/3C CC/CV charging and discharging method. A dimensionless capacity, Q^*, is defined as the ratio of capacity of the aged cell to that of the fresh cell. The charging time and dimensionless capacities of the several charging protocols for comparison are plotted in Figure 6 a) and b), respectively. For CC/CV charging, increased C-rate reduces the charging time, however accelerates the capacity fade. Compared with the charging protocol of CC/CV(0.75C), the designed FC protocol takes less charging time when battery is charged to 80% SOC, but needs more charging time when battery is charged to 100% SOC, as shown in Figure 6 a). The designed FC protocol decreases the capacity fade compared with the charging protocol of CC/CV(0.75C) because of the limitations of side reaction rate and ion concentration. However, compared with the charging protocol of CC/CV(0.33C), the the capacity fade fo the designed FC protocol is still too large, which might be caused by the lithium plating.



a) b)

Figure 6: Comparison of five charging protocols. a) Charging time; b) capacity fade.

3.2 Side reaction rate and lithium deposition reaction rate

Major three limitations are cutoff voltage, side reaction rate and the anode potential for lihtium plating. Charging C rates as a function of SOC are calculated using the ROM validated for a large format NMC/Carbon lithium ion cell with a capacity of 39Ah and plotted in Figure 7. When charged with 8C current rates from 0% SOC, lithium plating and side reactions will be accelerated. The first limitation during the low SOC is the anode potential that should not be lower than the zero voltage and then the side reaction rate and the terminal cutoff voltage that is 4.2V. In addition, the high charging current induces extreme high heat generation that should be rejected properly. Therefore, the provisional maximum charging current is set as 5C.

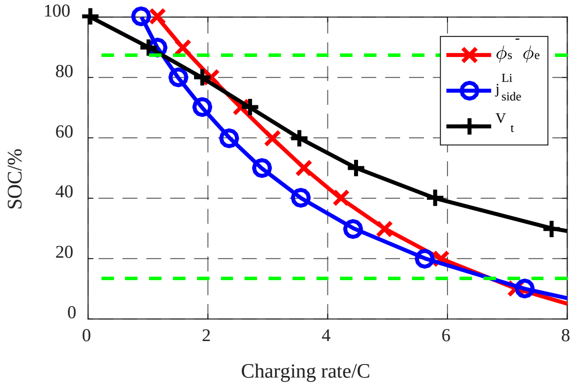
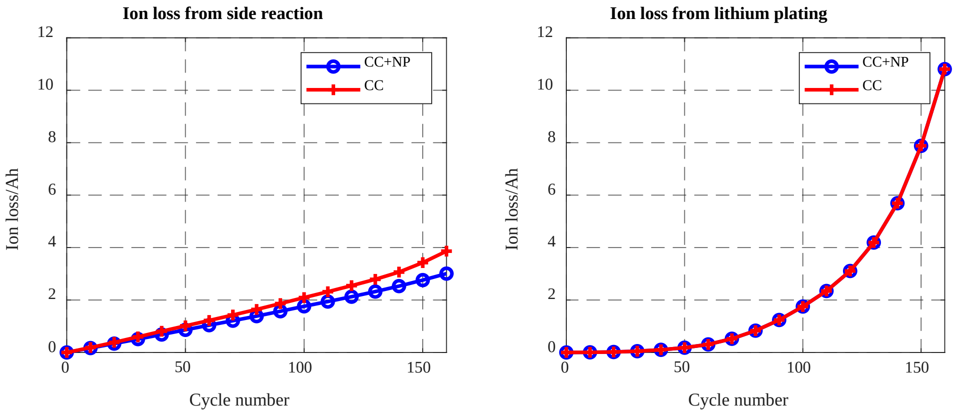


Figure 7: Different limitations for charging C rates as a function of SOC.

Theoretically, no lithium plating could be formed if the anode potential does not decrease beyond 0V. However, once lithium plating is formed for any reasons, further reaction is induced and accelerated. Therefore, preemptice measure is carried out with pulse current charging. Basically, the pulse charging methods bring several positive effects on degradation; 1) promote ions relaxation in electrodes to reach an equilibrium state of concentration of the ions, 2) reduce high ion concentration at surface and suppress formation of lithium plating, and 3) particularly, negative pulse (NP) recovers ions from plated lithium. The higher the magnitude of discharging pulse currents is, more effective is stripping [16].

Particularly, effects of negative pulse charging are investigated, where cycling tests are conducted using 5C CC charging with 2C negative pulse and CC charging with the same charging speed up to 160 cycles. Firstly, the capacity fade is compared with the one obtained from the ROM-EKF with degradation models and then ion loss and ion recovery from side reactions, lithium plating, and lithium stripping are separately caculated and plotted in Figure 8. As aforementioned, the side reactions occur from the beginning of cycling and ion loss increases lineary. On the other hand, the lithium plating occurs after 50 cycles and ion loss increases rapidly. In case of only CC charging, there is no lithium stripping, so capacity gets fade rapidly, which is caused by the decreased volume fraction of electrode and electrolyte, which also leads to an increase of side reaction after 50 cycles. However, employment of the CC + NP charging, due to recovery of ions from the plated lithium, capacity fade becomes much smaller and the difference of capacity loss is approximately 25% at 160 cycles



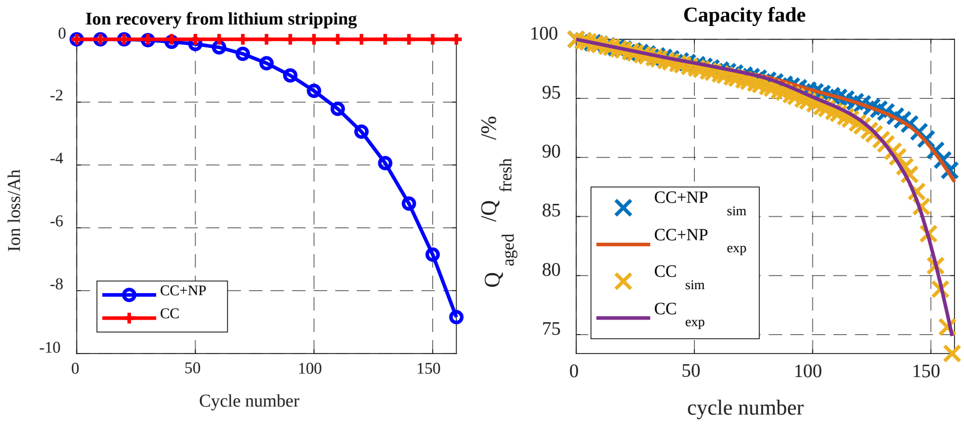


Figure 8 Ion loss and recovery from side reaction, lithium plating, and lithium stripping and capacity fade from experiment and simulation using CC and CC+NP charging

Adding NP during CC charging allows to minimize degradation by lithium stripping without increasing extra charging time. The proposed charging algorithm combined with CC and NP is called a fast charging method with negative pulse (FCNP). Current profiles of the FCNP at different cycles are plotted in Figure 9, where the charging current pulses at the low SOC range to suppress lithium plating and promote lithium stripping and decreases liks stairs to minimize the side reactions. As cycle number increases, the charging time becomes longer because of the decreased volume fraction of electrode and electrolyte and increased internal resistances.

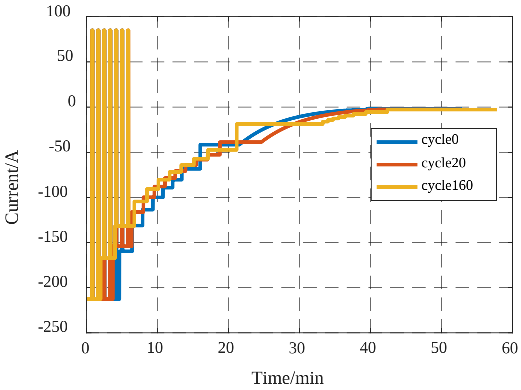


Figure 9: Current profile of the FCNP at 0, 20, and 160 cycles.

The FCNP is implemented in BIL and its charging time and capacity are measured and compared with those of 2C and 3C CC/CV charging protocols. The charging times at different SOC intervals plotted in Figure 10 show that the charging time by FCNP up to 40% SOC becomes 50% less than that by 2C CC/CV and 31% less than that by 3C CC/CV because of the high charging current at low SOC range. When charged up to 60% SOC, the charging time by FCNP becomes 43% less than that by 2C CC/CV, and 18% less than that by 3C CC/CV. However, because of low charging current at high SOC range, the charging time after 80% SOC by FCNP takes longer than that by 3C CC/CV.

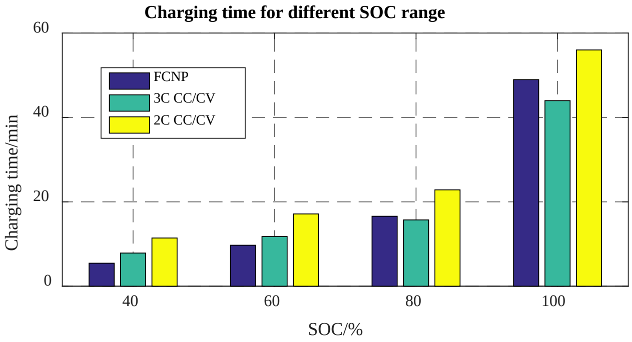


Figure 10: Charging times at different SOC intervals using FCNP, 2C CC/CV, and 3C CC/CV.

The measured and simulated capacity loss are plotted in Figure 11 as a function of cycle number. Because 3C CC/CV charging protocol exceeds limitations of charging currents set for lithium plating and side reaction, degradation is accelerated and the loss of the capacity becomes larger as the cycle number increases. From the beginning of cycling, the capacity loss by 3C CC/CV charging is always greater than that by FCNP and 2C CC/CV charging and decreases rapidly after 30 cycles by the cycles when lithium plating starts. The proposed FCNP has shown significant improvements in preventing the capacicy loss. The capacity loss is almost the same as that by 2C CC/CV charging and approximately 23% less than that by 3C CC/CV charging at 60 cycles.

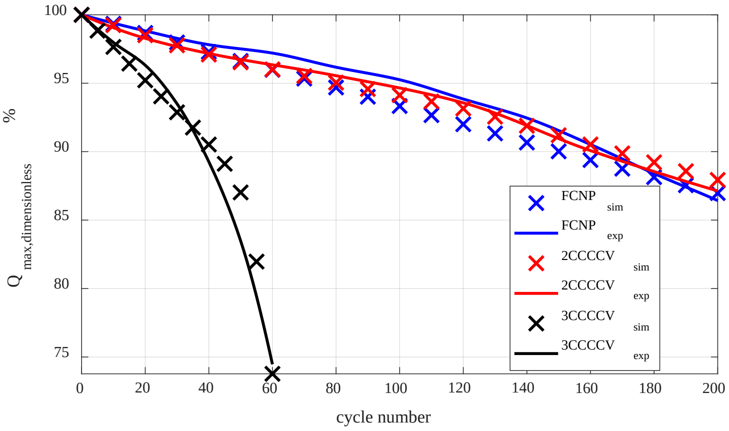


Figure 11: Capacity loss by FCNP, 2C CC/CV, and 3C CC/CV.

3 Commerciazation possibilies

The proposed method can be implemented for commercial products with several ways. Firstly, specifications for a battery system and charger should be known and then a charging protocol can be extracted from BIL using the ROM with EKF along with the degradation model. Currently, the ROM that embeds side reactions, lithium plating and stripping is available. Once the specifications of the battery system are given, the ROM parameters at BOL is determined using a genetic algorithms, while those at MOL and EOL are automatically updated by advanced parameter estimation methods. Finally, two solutions are possible, based on extracted data or ROM-EKF. The former requires less hardware performance, but might be less robust againt any change of parameters, while the latter does more performance of the hardware and is efficient until EOL.

4 Conclusion

A new fast charging method is proposed that suppreses the main causes for degradation while the charging time can be reduced. The internal physical variables primarily responsible for side reactions and lithium plating and stripping are derived from a reduced order electrochemical model that facilitates an implementation into a real time system. In addition, EKF is employed to minimize influences of noise on measurement and to estimate states such as ion concentrations and potentials. The proposed fast charging (FCNP) includes negative pulse charging current to suppress the lithium plating and promote recovery of ions out of plated lithium. The charging method has been tested experimentally by battery-in-the-loop system. The results have shown that the charging time by FCNP is approximately 50% less than that of 2C CC/CV charging for the interval from 0% to 40% SOC and 43% for the interval 0% to 60% SOC, while 31% and 18% less than that of 3C CC/CV charging for the interval from 0% to 40% SOC and 60% SOC, respectively. At the same time, the capacity loss by FCNP is comparable to that by 2C CC/CV charging, which is approximately 23% less than that by 3C CC/CV charging after 60 cycles.

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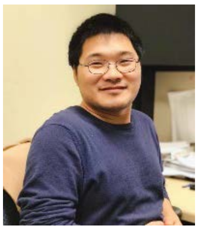
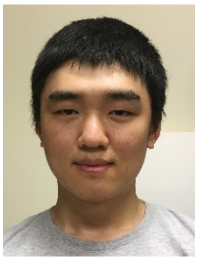
	Yilin Yin received his B.S. degree in Vehicle Engineering from China Agricultural University in 2010 and M.S. in Mechanical Engineering from Beijing Institute of Technology in 2013. He is currently pursuing PhD degree in Mechanical Engineering, Auburn University, where his research efforts focus on development of electrochemical thermal model of lithium-ion battery and its application for design of fast charging algorithm.
	Minseok Song received his B.S. degree in Mechanical engineering and Green energy engineering in 2015 from Yeungnam University, Gyeongsan, Korea and M.S. in Mechanical engineering in 2017. He is working towards his Ph.D at the mechanical engineering in Auburn University, USA. His research focuses on development of electrochemical and thermal model of lithium-ion battery considering degradation and its application for fast charging methods.