SAFE TEMPERATURE CONTROL OF LITHIUM ION BATTERY SYSTEMS FOR HIGH PERFORMANCE AND LONG LIFE

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by
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Abstract

Lithium-ion (Li-ion) batteries have diverse applications such as portable electronics, energy storage, hybrid electric vehicles (HEVs), plug-in HEVs, and electric vehicles (EVs). High energy density and longer life are major reasons for the widespread use of lithium-ion batteries. Presently, almost all major automobile manufacturers have hybrid vehicles in the market. HEVs can also help to reduce greenhouse gases and improve powertrain efficiency, but Li-ion battery life and performance significantly depend on the operating temperature and usage. High battery temperature increases battery degradation but this study proposes a counter-intuitive hypothesis that the life of lithium ion power cells can be increased by judiciously increasing the battery temperature in high power applications such as HEVs. End of life (EOL) in power applications often defined as when the battery is no longer able to provide the required charge/discharge power because the battery voltage exceeds the maximum/minimum allowable voltages associated with the battery’s chemistry. It is experimentally shown that battery life can be increased by step-wise temperature increases whenever the battery voltage exceeds a voltage limit.

The temperature and capacity of individual cells affect the current distribution in a battery pack. Non uniform current distribution among parallel-connected cells can lead to capacity imbalance and premature aging. This study develops models
that calculate the current in parallel-connected cells and predict their capacity fade. The model is validated experimentally for a nonuniform battery pack at different temperatures. The study also proposes and validates the hypothesis that active temperature control can reduce capacity mismatch in parallel-connected cells. Three Lithium Iron Phosphate (LFP) cells, two cells at higher initial capacity than the third cell, are connected in parallel. The pack is cycled for 1500 HEV cycles with the higher capacity cells regulated at 40°C and the lower capacity cell at 20°C. As predicted by the model, the higher capacity and temperature cells age faster, reducing the capacity mismatch by 48% over the 1500 cycles. A case study shows that cooling of low capacity cells can reduce capacity mismatch and extend pack life.

Lithium ion cells are increasingly being used in high power applications. There are four battery characteristics that are interlinked: Battery life, capacity, operating temperature, and usage. The goal of battery pack design is to minimize the battery pack cost or to maximize the battery pack life or both if possible. In this study, a model based process is developed that selects battery operating temperature and capacity to optimize the life and cost of the battery pack under prescribed usage.

At high temperatures, battery degradation increases and reduces battery life, but battery internal resistance reduces and improves battery performance. Batteries have a maximum allowable voltage limit based on degradation minimization, so the battery capacity is selected large enough to stay within the limit over the entire life of the pack. A real-time control algorithm is developed to vary the temperature of cells to improve their charge acceptance and reduce HEV pack size while maintaining battery life. The proposed algorithm has two strategies. First, the battery pack temperature is increased when its state of charge (SOC)
is high because the cell is more likely to exceed maximum voltage limit at high SOC. Second, the battery pack temperature is increased if a high current pulse is expected because higher cell temperature reduces the internal resistance and the corresponding voltage swing.

Besides battery performance, battery safety also has a paramount importance. Battery internal short and overcharge are two dangerous abuse conditions which can lead to the catastrophic results such as fire, smoke, or thermal runaway. Any accident related to battery systems pushes back the acceptability of new technology. This study explores the battery nail penetration and overcharge tests under different conditions. Internal short circuit occurs when a direct current path within a battery is established. A nail penetration test is used to simulate the internal short circuit process, which involves penetrating a test cell/pack with an electrically conductive nail. Gathering useful data at the point of penetration during nail penetration tests is very challenging due to the inherently destructive nature of the test. An intelligent nail (iNail) design is developed to conduct battery cell and pack level nail penetration tests. A prototype stainless steel iNail is manufactured and presented. Multiple thermocouples are placed inside the iNail. The iNail successfully recorded the temperature time history around the penetration point during the nail penetration test of a 4Ah pouch cell. Battery overcharge tests of the cylindrical cells are performed for 2 different chemistries NCA (lithium nickel cobalt aluminum oxide) and NCM (lithium nickel manganese cobalt oxide). Fresh cells and aged cells are overcharged at 1C and 10C rate at 45°C and -20°C.
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List of Symbols

\( a_s \)  active surface area per electrode unit volume, cm\(^2\)/cm\(^3\)
\( A \)  electrode plate area, cm\(^2\)
\( c \)  concentration of Li in a phase, mol/cm\(^3\)
\( D \)  diffusion coefficient of Li, cm\(^2\)/s
\( E_{act} \)  activation energy, J/mol
\( F \)  Faraday’s constant, 96,487 C/mol
\( I \)  applied current, A
\( i_0 \)  exchange current density of electrode reaction, A/cm\(^2\)
\( j^{Li} \)  reaction current resulting in production or consumption of Li, A/cm\(^3\)
\( L \)  cell width, cm
\( Q \)  capacity, Ah
\( r \)  radial coordinate within active material particle, cm
\( R \)  universal gas constant, 8.3143 J/mol K
\( R_c \)  contact resistance, \( \Omega \)cm\(^2\)
\( R_{ct} \)  charge transfer resistance, \( \Omega \)cm\(^2\)
\( R_s \)  radius of active material particles, cm
s  Laplace transform variable
T  absolute temperature, K
  time, s
U  open circuit, or equilibrium potential of an electrode reaction, V
V  voltage, V
  transference number of Li\(^+\) with respect to solvent velocity
  negative electrode solid phase stoichiometry and cell width spatial coordinate, cm
y  positive electrode solid phase stoichiometry

Greek symbol
  \(\alpha_a, \alpha_c\) anodic and cathodic transfer coefficients for an electrode reaction
  \(\varepsilon\) volume fraction or porosity of a phase
  \(\eta\) overpotential of an electrode reaction, V
  \(\kappa\) Li\(^+\) conductivity of electrolyte, S/cm
  \(\kappa_D\) Li\(^+\) diffusional conductivity of electrolyte, A/cm
  \(\sigma\) e\(^-\) conductivity of electrode solid matrix, S/cm
  \(\phi\) phase potential, V

Subscript
  \(e\) electrolyte phase
  \(max\) maximum value
  \(n\) negative electrode
  \(p\) positive electrode
  \(ref\) reference value
  \(avg\) average

xvii
s separator

s, e solid electrolyte interface

sei solid electrolyte interface layer

0% zero state of charge reference state

100% 100% state of charge reference state

Superscript

eff effective

Li Li lithium species

+ ion
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Dedicated to my wonderful parents
Chapter 1 | Introduction

Improving the fuel efficiency and reducing the emissions are major goals of the automobile industry. Transportation accounts for 28% of the nation’s greenhouse gas emission and 59% of the nitrogen oxide emission [3]. Currently battery market is expanding rapidly with the annual growth of 25% [4], and EVs and HEVs can help to achieve higher fuel efficiency and reduced emission goals [5].

1.1 Lithium Ion Battery Performance and Aging

Battery management system (BMS) requires fast and reliable battery models to control the battery packs for the optimal usage and safety. Equivalent circuit models are most commonly used in the BMS because they are computationally efficient for the real time implementation [6–9], but these models lack the physical understanding of the underlying process. They also require extensive experiential data to tune for different capacities and temperatures. On the other hand, full order models have a better physical understanding but they are not fast enough to implement in real time [10–13]. As an alternative, physics-based reduced order models can be used because they are computationally efficient and have a good physical understanding [1, 14–19]. Prasad et al. [15, 20] developed a linear single
particle, performance model that could be realized as an equivalent circuit, unifying these two approaches. Tanim et al. [16] developed an Enhanced Single Particle Model (ESPM) by adding electrolyte dynamics to the single-particle model. These models are developed by simplifying the full order models under certain assumptions, but they are sufficiently accurate and fast enough to implement in real time.

1.1.1 Performance Model

Figure 1.1 shows a schematic diagram of the pseudo-2D Li-ion battery model [10–13] that is the basis of the theoretical study. The current input $I(t)$ charges ($I > 0$) and discharges ($I < 0$) the cell to produce the output voltage $V(t)$. Lithium ions shuttle back and forth between the negative electrode ($x < L_n$) and positive electrode ($L_n + L_s < x < L_n + L_s + L_p$) through the separator ($L_n < x < L_n + L_s$). The model variables include solid phase concentration $c_s$ in the active material particles, electric potential $\phi$ in the solid (s) and electrolyte (e) phases, electrolyte phase concentration $c_e$, and exchange current density $j^{Li}$ and over-potential $\eta$ in the two electrodes. Performance and aging models given in Appendix-A are the basis of this study. Table A.1 shows the governing equations of the Enhanced single particle model (ESPM), reduced order model developed in [16]. The following assumptions apply to the ESPM model:

1. The ESPM assumes that $j^{Li}$ is uniform (infinite electrode conductivity) across each electrode so all material particles are in parallel and the solid phase of each electrode can be represented by a single, spherical material particle.

2. The battery temperature $T$ is assumed to be fixed and uniform.

3. The ESPM is linearized around a fixed SOC set point and current inputs are assumed to be small.
These assumptions reduce the full-order model to the simplified transfer functions given in Table A.2. The solid phase Li-ion diffusion Eq. (A.1) is solved using Laplace transformation and Padé approximation resulting in the transfer functions for the positive and negative particles in Eqs. (A.8a) and (A.8b) [21]. Equation
A.2 governs the solid phase charge conservation and reduces to uniform current distributions within the two electrodes in the Eqs. (A.9a) and (A.9b). Equation (A.3) and (A.4) govern the electrolyte dynamics and are simplified using a third order Integral Method Approximation to obtain the second order transfer function in Eq. (A.10). The Butler-Volmer (B-V) relating current density and overpotential is linearized in Eq. (A.11a) and (A.11b). The overpotential at the particle surface in Eq. (A.6) and output voltage Eq. (A.7) become Eq. (A.12) in the ESPM. \( R_c \) in Eq. (A.12) is the contact resistance of the circuit which varies depending on the end connections and needs to be tuned for each different experiment. The parameters for the ESPM model of the Lithium ion phosphate (LFP) cells are given in Table A.3.

1.1.2 Aging Model

According to USCAR, the EOL goal of an HEV is 15 years and for an EV and PHEV, it is 10 years \cite{22}. To achieve this goal, efficient battery degradation models are required to predict the battery capacity fade and impedance rise in different conditions.

LFP batteries have a stable cathode due to its olivine crystal structure. A major degradation mechanism in LFP batteries is the solid electrolyte interphase (SEI) layer growth on the graphite negative electrode that leads to the loss of the active material and capacity \cite{23–25}. Safari et al. \cite{26} experimentally showed that SEI layer growth is affected by usage and operating temperature. Ramadass et al. \cite{27} proposed a simplified, control-oriented SEI growth model. Randall et al. \cite{28} simplified this model and proposed an iterative approach to calculate the SEI side reaction current density. Tanim et al. \cite{2} further simplified the aging model to develop an analytical formula for the capacity fade.
In this study, the model developed Tanim et al. [2] is used and further validated for different temperature and capacity. The model assumes that the current density of the aging side reaction is much less than the current density of main charge/discharge reaction and the resistance growth due to SEI layer formation is negligible. The governing equations and model parameters for calculating the capacity fade are given in Table A.4 and A.5, respectively. The side reaction current density \( j_{sei} \) is related through Tafel kinetics to the side reaction overpotential \( \eta_{sei} \). Side reaction equilibrium potential, \( U_{sei}^{ref} \), is 0.4 V. The overpotential depends on the fields in the negative electrode \( \phi_n \) and the electrolyte \( \phi_e \) calculated from the performance model. The capacity loss \( Q_{loss} \) is proportional to the time integration of \( j_{sei} \).

Temperature plays a critical role in the performance and aging of Li-ion batteries. High temperatures will accelerate the battery aging [26], but reduce the impedance of the battery pack [29] that enhances the battery performance. Figure 1.2 shows that higher battery temperature reduces the battery internal resistance, which will reduce the maximum battery voltage and increase the voltage-based EOL. However, a higher temperature will accelerate the aging and decrease the state of health (SOH) based EOL. There exists an optimal battery temperature for different conditions [29].

Lithium plating is another degradation mechanism which is predominant only at the lower temperature [30–32]. Waldmann et al. [33] quantitatively analyzed the effects of temperatures on the aging behavior of the lithium ion batteries. It is experimentally shown that there are two different aging mechanisms for the ranges of -20°C to 25°C and 25°C to 70°C. Below 25°C, the aging rate increases with decreasing temperature due to lithium plating, while above 25°C aging rate increases with increasing temperature due to SEI layer growth. Lithium plating
occurs when the side-reaction overpotential is negative. Perkins et al. [34] developed a lithium plating model to estimate the capacity loss and resistance rise by creating a simplified approximation to a model proposed by Arora et al. [35].

Wang et al. [36, 37] developed all condition battery (ACB) for the low-temperature battery operation in EVs. ACB battery has an internal heater that can use the battery’s own energy to heat itself efficiently to avoid lithium plating during extreme cold environments [38, 39]. Yang et al. [40] developed the innovative self-heating Li-ion battery (SHLB) with the focus of battery heating speed. Conventional Li-ion cell heating strategies either internal heating via battery’s resistance and external heating via resistive heaters are slow and inefficient. SHLB achieves nearly 100% heating efficiency and 40 times shorter heating time.

Figure 1.2. Battery internal resistance and degradation rate variation with the battery temperature.
1.1.3 Battery Pack Balancing

Battery pack life and performance is also affected by cell imbalance. Cell imbalance can happen due to internal sources as manufacturing variation, impedance variation, and different self-discharge rate or external sources as different temperature across cells [41]. Cell imbalance may cause voltages to differ significantly and create unsafe conditions.

Fixed [42] and switching [43] shunt resistance methods use a shunt resistance in parallel to the cell to divert the extra current to the resistance and protect the cell from overcharge. The major drawback of the shunt resistance is the wastage of energy dissipated as heat in the shunt resistance while there are some cells in the string that can absorb extra energy. This wastage can be minimized using active balancing methods such as switch capacitor [44], single switch capacitor [45], and double tired capacitor [46,47]. These methods use the combination of capacitors to transport the energy from high energy cell to low energy cell.

1.1.4 Battery Temperature Modeling

Temperature monitoring is required to avoid battery overheat as it may lead to disaster. Usually, battery cell surface temperature can be measured directly, but battery internal temperature is inaccessible until the battery is designed especially with internal temperature sensors [48,49]. Kim et al. [50] developed a computationally efficient thermal model to estimate the radical distribution of the temperature in a cylindrical cell. The model is validated for LFP batteries and further extended for the estimation of the temperature distribution under unknown cooling conditions [51,52]. Lin et al. [53] developed a two state thermal model for battery core temperature estimation. Online parameters estimation scheme is designed for the cylindrical batteries. The model is able to estimate
battery core temperature even with variable heat generation due to variable internal resistance. The model is validated for cylindrical LFP batteries [54]. Lin et al. [55] also developed the thermal model for battery clusters which includes cell to cell heat conduction and thermal dynamics of the coolant flow due to convection. Later, the thermal model is coupled with battery equivalent circuit model to develop a computationally efficient 5-state model [56]. The model captures the state of charge (SOC), terminal voltage, surface, and core temperature of a battery for a wide range of operating conditions.

1.2 Battery Safety

1.2.1 Battery Internal Short Circuit

Higher energy density in large format lithium-ion batteries brings major safety concerns [57]. The safe and efficient operation of lithium-ion batteries are critical for the wider acceptability of the battery technology including vehicle electrification. Events of the fire in mobile phone and electric vehicles hinder the growth of the battery technology [58,59].

The internal short circuit is the most dangerous abuse scenario which occurs when a direct current path within a battery is established. A lot of current passes through the short circuit, which produce large amounts of heat. As a result, self-sustaining exothermic reactions are triggered which finally lead to thermal runaway. A pinch test can be used for internal short circuit [60]. Usually, a nail penetration test is used to simulate the internal short circuit process, which involves penetrating the test cell with an electrically conductive (e.g. stainless steel) nail [61]. Pass or failure of the cell is determined by the presence of smoke or flame following the nail penetration. Zhao et al. [62,63] developed a numerical model for the battery nail
penetration. Effect of shorting resistance, nail diameters, nail thermal conductivity, the number of shorted layers, and cell capacity is analyzed for the maximum battery temperature and temperature distribution.

It is crucial to obtain useful data during nail penetration tests to understand the dynamics of the cell during penetration. Gathering useful data during nail penetration tests is very challenging due to the inherently destructive nature of the test and the limitation of using more sensors with the conventional solid nail design [64–67]. Hatchard et al. [68] design a smart nail where a thermocouple is welded inside a copper nail tip to measure temperature precisely at the puncture point. Nail penetration tests with the smart nail require precise control of the nail position to position the nail tip inside the cell.

1.2.2 Battery Overcharge and Overdischarge

Battery overcharge is an unsafe condition as it can cause thermal runaway. Tobishima et al. [66,69] showed that at high current the overcharge is more catastrophic. It was observed that at charge rates of 1 C and 1.5 C, batteries swelled but the safety vent did not open or smoke. However, batteries overcharged by more than 2 C caught fire after the rapid cell temperature increase caused by the electrolyte decomposition. Leising et al. [70,71] showed that the point of cell rupture during overcharging is related to the amount of lithium in the cathode. When lithium ends in the cathode, different exothermic side reactions occur. Doughty et al. [72] also explained the same result that the thermal runaway during charging is related to the amount of Li available in the cathode of different chemistry. Belov et al. [73] did the overcharge experiment using CC-CV charging method and called it ”Soft” overcharge. To avoid the overcharge and safe control of the battery, correct SOC estimation is desired. Plett [74–76] introduced the use of Kalman filtering for
SOC estimation using equivalent circuit models.

Maleki et al. [77] showed the effect of the over-discharging on cell capacity, impedance, and thickness. Cells were overcharged and kept at different voltages. Overdischarge to 2.0 and 1.5 V had small effects on cycle-life, but over discharge to 1.0 and 0.5 V led significant capacity loss. Also, over discharge to 0 V shows high swelling after the third cycle of charging and over discharging. Xiong et al. [78] used 1Ah A123 18650 cells for failure detection due to over discharging and observed no failure during the over discharging phase, but all failures in the recharge phase.

1.3 Overview of the Dissertation

Li-ion batteries have higher energy density and longer life that make them suitable for wide range of applications. HEVs have better gas mileage due to the efficient storage of the regenerative energy [79], but battery operating temperature and usage affect its life that can increase the cost of operation for EVs and HEVs. This study explores the safe temperature control strategies for improving lithium ion battery performance and life. This is the first study to increase the battery temperature judiciously for battery performance and life improvement in HEVs.

In chapter 2, a counter-intuitive hypothesis that increasing the battery temperature can increase the battery life in HEVs is proposed and validated experimentally. Maximum/minimum voltage limits based battery EOL is considered in this study. Increasing the battery temperature will accelerate the battery aging but it will also reduce the battery internal resistance and output voltage swing. The battery pack is operated at its normal temperature until EOL. Then the battery temperature is increased to reduce impedance and allow cycling without exceeding voltage limits, thus prolonging EOL.

In chapter 3, a hypothesis that the temperature control can reduce the battery
pack capacity mismatch is presented and validated experimentally. Current
distribution in the cells connected in parallel depends on the capacity and
temperature of the individual cell. Cells at a higher temperature or with higher
capacity will provide more current and degrade faster. So it is possible to balance
the battery pack by temperature control.

While validating the experimental results, the aging model is validated at higher
temperatures, different initial SOHs, and wider ranges of SOH. The ESPM model
is further extended to predict the current distribution among different capacity
cells connected in parallel at a different temperature. In chapter 4 and 5, these
comprehensively validated models are used for designing the battery pack size and
operating temperature profile.

In chapter 4, a process is developed to select the battery pack capacity for
different applications to minimize the battery cost while maintaining the battery life.
Specifically, this chapter answers following questions: How does one select battery
pack size to minimize the cost of operation for a specified battery temperature and
usage? How does one select battery temperature to maximize the battery life for a
specified battery pack size and usage? How does one select battery pack size and
operating temperature for a specified battery life requirement and usage?

In chapter 5, strategies are developed for real-time temperature control of
HEV batteries to improve battery charge acceptance and reduce battery pack size.
Specifically, this chapter explores two strategies to improve the battery charge
acceptance. The first strategy is to increase the battery temperature at high SOC
and second strategy is increase the battery temperature at high current pulses.
Simulation results predict battery pack size reduction of up to 30% by active control
of the battery temperature,

In chapter 6, battery safety during nail penetration and overcharge is discussed.
An iNail is designed for nail penetration tests. Different sensors like thermocouples and strain gauge can be placed inside the iNail which improve the quality of data in nail penetration tests. Battery overcharge tests are also performed for NCA and NCM chemistry under different conditions.

Experiments and simulations are performed for LFP batteries. Results would be transferable to other chemistries to an extent. The ESPM model works for other chemistries such as NCM and NCA though the model needs to be tuned for different cells. Other chemistries will also have SEI layer growth on the negative electrode, but they might also have other degradation mechanisms.
Chapter 2  
Elevated Temperature for Life Extension of Lithium Ion Power Cells

2.1 Introduction

According to the U.S. Advanced Battery Consortium (USABC), battery EOL in EV applications is reached when battery remaining capacity or peak power decreases to 80% of the rated value [80]. The USABC definition of EOL may not be suitable for HEVs. HEVs operate on charge sustaining cycles in small SOC windows to provide the required power without exceeding pre-defined voltage limits. An alternative EOL definition for HEVs is proposed in FreedomCAR Battery Test Manual [81] as follows: A battery pack is at EOL when the voltage first exceeds a maximum/minimum voltage limit during normal operation and the test profile cannot be executed within the voltage limits. The maximum allowable voltage is determined by the battery chemistry and manufacturer to maintain safe and long-life operation. This EOL definition is not explicitly based on SOH. HEV
batteries can be designed to provide the required pulse power after 80% SOH. EOL implicitly depends on SOH, however, and the impedance rise associated with aging. Normal operation is defined by reasonable charge/discharge current inputs that are represented by a charge-sustaining duty cycle associated with typical HEV operation. Finally, it is typically charge acceptance during breaking that is most likely to result in exceeding a voltage limit so the maximum voltage limit often dictates EOL.

This study shows that HEV EOL and battery life can be extended by judiciously increasing the battery temperature. HEV battery pack often have thermal management systems so raising the temperature may simply entail changing set point in the controller. The battery is operated at its normal temperature until EOL. Then the battery temperature is increased to reduce impedance and allow cycling without exceeding voltage limits, thus prolonging EOL. Higher battery temperature increases the battery degradation rate [29], however, so the battery capacity and impedance degrade at the accelerated rate until the battery exceeds voltage limit once again. The process of increasing battery temperature in response to exceeded voltage limits can be continued to provide more cycles. Eventually, the battery temperature will reach the safety limit and the battery will truly be dead. This study is the first to put forth the counter-intuitive hypothesis that heating an HEV battery can extend its life. The hypothesis is theoretically and experimentally proven for LFP batteries.

### 2.2 Experimental Setup

Figure 2.1(a) shows the experimental setup used to validate the proposed models and provide evidence that supports the hypothesis. Six LFP cells with
Figure 2.1. Experimental testing of LFP battery pack with 3 cells in parallel: (a) Set up, and (b) Procedure.

different initial SOHs are aged in two different experiments. In both experiments, a pack comprising three commercially available 4.5 Ah cylindrical graphite/LFP cells connected in parallel is placed in the temperature controlled environmental chamber. The packs are cycled using an HEV profile with an Arbin BT-2000. The three cells have the same voltage. The current of the individual cells is measured using Hall Effect current sensors. Cell capacity is measured at specific intervals in four steps: (1) Charging to 3.6 V at 1C constant current; (2) Holding voltage
Figure 2.2. Experiment 1 cycling results: (a) Theoretical (lines) and experimental (symbols) capacity fade, (b) Model error versus time, and (c) Battery mean temperature of each cycle.
constant at 3.6 V until the current drop to C/20; (3) Resting for 30 min; and (4) Discharging at 1C rate to a cut-off voltage of 2.0 V.

**Experiment 1: Model Validation** Cell 1, cell 2, and cell 3 have initial capacities of 4.41 Ah, 4.41 Ah, and 3.98 Ah, respectively. Cell 1 and Cell 2 are fresh cells, and cell 3 is 10% aged from previous experiments and calendar aging. Initial SOC of the cells is 44.55%. The pack is cycled using 4200 seconds long charge sustaining HEV cycle that has 10.5 % SOC swing and 182 Amp and -195 Amp maximum and minimum current, respectively, for 1250 cycles (approximately 3 months) at 37°C environmental chamber temperature.

**Experiment 2: Life Extension** Cell 4, cell 5, and cell 6 in the second pack have initial capacities of 3.96 Ah, 4.07 Ah, and 4.11 Ah, respectively. All three cells have been aged in other experiments and/or calendar aged. Initial SOC of the cells is 44.55%. In this experiment, the voltage limit is defined to be 3.571 V and 2.00 V, narrower than required by LFP chemistry to accelerate EOL. The environmental chamber temperature starts at 37°C. The pack is cycled using the SOC resetting protocol in Fig. 2.1(b) until EOL at 250 cycles. The SOC of individual cells is reset every 4 days (around 75 HEV cycles) to account for the slow SOC drift associated with Arbin current noise. At EOL the pack can no longer provide the required charge acceptance without exceeding the maximum voltage limit. The battery pack temperature is then raised by 1°C and it is once again able to perform the HEV cycle without exceeding the voltage limits. At 1250 cycles, the battery pack again reaches EOL, and its temperature is raised by 1°C so that it cycles further. In this way, chamber temperature is increased up to 40°C with stepwise temperature increases of 1°C. Overall, the battery pack undergoes 2500 cycles for approximately 6 months.
2.3 Results and Discussion

2.3.1 Experiment 1: Model Validation

Figure 2.3(a) shows the voltage time response of a cell to the partial cycle current profile in Fig. 2.3(b). The model-predicted voltage matches the experimental results within ±0.5%, validating the performance model. Figure 2.2(a) shows the theoretical and experimental capacity fade results for all six cells. The temperature histories are shown in Figure 2.2(c) and vary from 39° C to 43° C. The experiment capacity fade and model predictions are shown for different initial SOH and temperature histories. Figure 2.2(b) shows the capacity fade prediction error is less than ±1% for all 6 cases validating the aging model.

2.3.2 Experiment 2: Life Extension

Figure 2.3 shows experimental results from experiment 2. Fig. 2.3(a) shows the voltage response, and experimental voltages match closely with the simulation voltages. The mean error was 9mV. During each cycle, there is one point with maximum voltage, typically after long and high current charge pulse as shown in Fig. 2.3(a) - (d). The minimum voltage is well above the limit. Thus, EOL for this cell, average SOC, and current profile is determined by the charge acceptance/maximum voltage limit. The SOCs of the individual cells drift upward over time and are manually reset every 4 days or roughly 75 HEV cycles. SOC drift causes the battery voltage to increase so only battery voltages shortly after SOC resets are used to determine whether voltage limit has been exceeded. Figure 2.4 shows the temperature profile for the first few HEV cycles after a SOC reset. The environmental chamber and cell temperatures are transient for a few cycles.
Figure 2.3. Experiment 2 cycling results: (a) Theoretical (green-dashed) and experimental (blue-solid) voltage response over 200 seconds of the HEV cycle that includes the maximum voltage (red-dot), (b) Current input corresponding to the voltage response in (a), (c) Voltage response during an entire cycle (d) Current profile of an entire cycle.
Temperature also influences voltage so we use the mean of the maximum voltages in HEV cycles 6, 7 and 8 after SOC reset as the $V_c$ to be compared with voltage limits. Figure 2.4 shows the temperature transients in the chamber and cells setting out after 4-5 cycles so cycle 6-8 are at steady state.

Figure 2.5(b) shows the complete data for experiment 2, consisting of maximum voltage for 2500 HEV cycles. The inset to Fig. 2.5(a) zooms in to help to explain the effect of the SOC drift on the maximum voltage. The marked points in the figure are as follows:

**Point A** - After every 75 cycles (~4 days of aging), the mean SOC has drifted by 4-5%, and the SOC is reset. Point A represents the maximum voltage in the last HEV cycle before SOC of the battery pack was reset.
Figure 2.5. Experiment 2 cycling results: (a) Maximum voltage in each cycle for 2500 cycles; $V_c$, the mean of the maximum voltage of cycles 6, 7 and 8 after SOC reset; and EOL, end of life voltage limit; (b) Cell and thermal chamber temperatures
Point $B$ - Point $B$ is the maximum voltage in the first HEV cycle after the SOC reset. The voltage at the point $B$ is lower than the voltage at the point $A$ because the SOC at point $B$ is lower than the SOC at the point $A$.

Point $C$ - Point $C$ is at $V_c$, the average of the maximum voltage in cycles 6, 7 and 8 after the SOC reset.

Points $D$, $E$, and $F$ - Points $D$, $E$, and $F$ corresponds to points $A$, $B$, and $C$ but at the next SOC reset.

Figure 2.5(b) shows the cell and experimental chamber temperature responses. The cell temperatures track the chamber temperature except a few degree warmer due to internal cell heating. The temperature dropouts/noise are due to start up artifacts.

10% aged cell pack was chosen for experiment 2 and aged for additional 250 cycles at 37° C environmental temperature. After the 250 cycles, $V_c$ was 3.571 V. The maximum voltage limit for an LFP battery is 3.6 V, but 3.571 V is used for EOL in this study. The value of the voltage limit is not critical to demonstrate the effect of increase cell temperatures on voltage response.

After the first 250 cycles, the environmental chamber temperature was increased by 1° C, and the cell surface temperature also increased by around 1° C. Increased cell temperature reduces the effective battery resistance and reduces the maximum voltage in the HEV cycle. The maximum voltage no longer exceeded the EOL, and the battery pack provided more cycles. In the experiment, battery's life was extended by 1250 cycles due to this 1° C temperature increase. After 1250 additional HEV cycles, $V_c$ exceeds the EOL, and the battery pack is no longer able to accept the required charge without exceeding the voltage limit.

Again, the environmental chamber temperature was increased by 1° C to 39° C and the cell surface temperature also increased by 1° C. The battery pack was able
to provide an additional 925 HEV cycles before $V_c$ again exceeded the EOL. After 2400 total cycles, the environmental temperature was again increased by $1^\circ$ C to show that $V_c$ can be reduced, and the battery’s life can be further extended. Figure 2.5(b) shows decreases in $V_c$ near cycle 250, 1500, and 2400 due to temperature increases and slow increase of $V_c$ from cycle 250 to 1500 and 1500 to 2400 due to capacity fade. Figure 2.5(b) also shows the model predicted $V_c$ and it matches experimental $V_c$ quite well.

### 2.4 Conclusion

The study theoretically and experimentally proves the hypothesis that increasing battery temperature can extend battery life. Two $1^\circ$ C temperature increases extend the life of a 4.5 Ah LFP cell by 2000 charge sustaining HEV cycles. These small temperature increases allowed the cell to cycle continuously for an additional 3 months.
Chapter 3  |  Temperature Control To Reduce Capacity Mismatch In Parallel-Connected Lithium Ion Cells

3.1 Introduction

In this study, the ESPM model is extended to parallel-connected cells to calculate the current distribution in a battery pack. The model is augmented with SEI layer degradation to predict aging. The performance and aging predicted by the model are validated experimentally for parallel-connected cells of different capacities at different temperatures. Also, active temperature control for the reduction of battery pack capacity mismatch is studied through models and experiments. Finally, a case study is presented to investigate active temperature control that can simultaneously maximize the battery life and minimize the battery pack capacity mismatch.
3.2 Mathematical Modeling

3.2.1 Single Battery Performance Model

Table A.1 lists the fundamental governing equations of the pseudo-2D cell model. Four partial differential equations (solid phase charge conservation, solid phase ion conservation, electrolyte phase charge conservation, and electrolyte phase ion conservation) are coupled via the Butler-Volmer and overpotential equations. Reduced order battery models simplify these fundamental governing equations using assumptions and numerical model-order reduction tools. Reduced order models are accurate over a limited operating range, but they are computationally efficient.

Following Tanim et al. [16], we use the reduced order ESPM, a single particle model that assumes infinite solid phase conductivity and uniform temperature distribution across the cell. The model describes the voltage output for a single cell as follows:

\[
\frac{V_{out}(s)}{I(s)} = \frac{\eta_p(s)}{I(s)} + \frac{\partial U}{\partial \tilde{C}_{s,e}} \frac{\tilde{C}_{s,e}}{I(s)} - \frac{\eta_n(s)}{I(s)} - \frac{\partial U}{\partial \tilde{C}_{n,e}} \frac{\tilde{C}_{n,e}}{I(s)} + \frac{\Delta \phi_e(L, s)}{I(s)} - \frac{R_c}{A} \tag{3.1}
\]

Individual terms of the transfer function are described in Table A.2.

The coefficients of the transfer function depend on the battery electrochemical and physical properties. The electrochemical properties are temperature dependent and the Arrhenius equation,

\[
\psi = \psi_{ref} \exp \left[ \frac{E_{act,\psi}}{R} \left( \frac{1}{R_{ref}} - \frac{1}{T} \right) \right], \tag{3.2}
\]

is used for to calculate the temperature-dependent parameters. Table A.3 lists the
parameters for a single cell.

### 3.2.2 Battery Pack Performance Model

For a battery pack with three LFP cells connected in parallel, each cell has the same voltage $V_{\text{pack}}$ and the battery pack current,

$$I_{\text{pack}} = I_{\text{cell}_1} + I_{\text{cell}_2} + I_{\text{cell}_3},$$  \hfill (3.3)

is the sum of the cell currents. For a uniform pack, the cell currents are equal. In a non uniform pack, each cell has a different transfer function

$$G_{\text{cell}_1} = \frac{V_{\text{pack}}(s)}{I_{\text{cell}_1}(s)}, \quad G_{\text{cell}_2} = \frac{V_{\text{pack}}(s)}{I_{\text{cell}_2}(s)}, \quad G_{\text{cell}_3} = \frac{V_{\text{pack}}(s)}{I_{\text{cell}_3}(s)},$$

based on its capacity, parameters (e.g. internal resistance), and temperature.

The individual cell current transfer functions can be calculated as:

$$\frac{I_{\text{cell}_1}(s)}{I_{\text{pack}}(s)} = G_T(s) G_{\text{cell}_1}(s),$$  
$$\frac{I_{\text{cell}_2}(s)}{I_{\text{pack}}(s)} = G_T(s) G_{\text{cell}_2}(s),$$  
$$\frac{I_{\text{cell}_3}(s)}{I_{\text{pack}}(s)} = G_T(s) G_{\text{cell}_3}(s),$$  \hfill (3.4)

where, $G_T(s) = \left( \frac{1}{G_{\text{cell}_1}} + \frac{1}{G_{\text{cell}_2}} + \frac{1}{G_{\text{cell}_3}} \right)^{-1}$. 

26
Figure 3.1. Three cells connected in parallel are placed in the thermal chamber. Cell 1 and 2 are insulated and heated using an external heater. Thermal chamber is kept at $20^\circ C$. (a) Schematic of the experimental setup, (b) Battery pack with Cell 3 without insulation, (c) Cell insulation layer 1, (d) Resistive heating element layer around the cell, (e) Another insulation layer over the resistive heater, and (f) cross section of the insulated cells.

### 3.2.3 Battery Pack Aging Model

Anodic overpotential for the intercalation/de-intercalation reaction for the $i^{th}$ cell can be calculated using ESPM:

\[
\frac{\eta_i(s)}{I_{cell_i}(s)} = \frac{R_{ct}^n}{a_n^n A_n L_n}
\]
Figure 3.2. Experimental temperatures for the first 11 HEV cycles: Cell 1 (blue), Cell 2 (red), Cell 3 (black), and Thermal chamber (green) temperatures.

\[ \eta_i = \phi^n_s - \phi^e - U_n(C^n_{s,e}) \]

Overpotential for SEI growth side reaction is

\[ \eta^{sei} = \phi^n_s - \phi^e - U^{ref}_{sei} \]

Thus,

\[ \eta^{sei}_i = \eta_i + U_n(C^n_{s,e}) - U^{ref}_{sei} \]

where \( U^{ref}_{sei} \) is 0.4V. The side reaction current for SEI layer growth and capacity loss of the individual cell are

\[ j^{sei}_i = -a^n_s i_{0,sei} \exp \left( -\frac{\alpha^F_n F}{RT} \eta^{sei}_i \right), \]
\[ \frac{\partial QLOSS_i}{\partial t} = -j^{sei} A_n L_n, \]

where \( QLOSS_i \) is the capacity loss in the \( i^{th} \) cell due to the SEI layer growth.

### 3.3 Experiment Setup

Three LFP cylindrical cells are connected in parallel to construct the battery pack shown schematically and in a photograph in Figure 3.1. The battery pack is placed in a thermal chamber to control the environmental temperature. An Arbin cycler charges and discharges the battery pack using a predefined HEV cycle. The Arbin also measures the battery pack voltage and current. Cell 1 and Cell 2 are insulated and heated using a resistive heater, so they operate at higher temperature than Cell 3. The mean temperatures of Cell 1, 2, and 3 are 39.5°C, 42°C, and 23°C, respectively. Hall Effect current sensors measure the current in the individual cells. Figure 3.2 shows the temperature of different cells and environmental chamber for the first 11 HEV cycles. The system reaches steady state within a couple of 4200s long HEV cycles.

The battery pack is cycled for 1500 HEV cycles and capacity is measured roughly every 250 cycles. Cell capacity is measured as follows: (1) Charging to 3.6 V at 1C constant current; (2) Holding voltage constant at 3.6 V until the current drops to C/20; (3) Resting for 30 min; and (4) Discharging at 1C rate to a cut-off voltage of 2.0 V. The initial capacities of Cell 1, 2 and 3 are 4.27Ah, 4.27Ah, and 3.98Ah, respectively.
3.4 Experimental Results and Discussion

3.4.1 Current Distribution

Figure 3.3 shows the experimental and simulated current in Cell 2 and 3 at the beginning of the cycling and after 1300 HEV cycles. The theory and experiment match extremely well, indicating the accuracy of proposed performance and aging models. Table 3.1 shows the maximum error is around 1% for the aged cell.

As expected, the current distribution is very sensitive to cell temperature, with higher temperature cells taking more current. Table 3.1 shows the RMS currents in the three cells, Cell 1 and 2 are roughly 18°C warmer than Cell 3 and they take almost 33% more current. Even the small temperature difference of 2°C between Cells 1 and 2 results in 0.5A more current going to the hotter cell 2.
Figure 3.4. Experimental validation of the battery capacity fade model for cells (1-3) at mean temperatures of 39.5°C, 43°C, and 23°C and initial capacities of 4.27Ah, 4.27 AH, and 3.98Ah, respectively. Simulated capacity fade: Cell 1 (black solid line), Cell 2 (red dashed line), and Cell 3 (blue dotted line). Experimental capacity fade: Cell 1 (black upward-pointing triangle), Cell 2 (red diamond), and Cell 3 (blue downward-pointing triangle).

The current distribution is relatively insensitive to cell capacity variation. Figure 3.3 and Table 3.1 show almost identical current distribution at the beginning and end of the test, even though the cell capacities are much closer at the end of the test.

### 3.4.2 Capacity Fade

Figure 3.4 shows that the cell capacity loss mirrors the current distribution. The hottest Cell 2 degrades the quickest and the coolest Cell 3 maintains its capacity.
Figure 3.5. Experimentally measured cell capacities standard deviation versus number of cycles.

The model predictions of capacity are not as accurate as performance. This is partially due to the capacity measurement inaccuracies. Accurate aging prediction also requires consideration of alternative aging mechanisms (e.g. electrode cracking). This may explain why the aging model under predicts capacity loss.

3.4.3 Mismatch Reduction Through Different Temperature Control

Figure 3.4 shows that the rate of capacity fade for Cell 1 and 2 is much higher than the rate of capacity fade for Cell 3 due to their higher temperature. The battery pack used for the experiment is not uniform with cell capacity varying by as much as 7.45% at the beginning of the test. The non uniform temperature distribution causes differential aging so the battery pack becomes more uniform
Table 3.1. Summary of the test results

<table>
<thead>
<tr>
<th>Cell No.</th>
<th>Initial Values before cycling</th>
<th>Final values after 1300 HEV cycles</th>
</tr>
</thead>
<tbody>
<tr>
<td>Capacity (Ah)</td>
<td>1 4.27</td>
<td>4.27</td>
</tr>
<tr>
<td></td>
<td>2 4.27</td>
<td>4.00</td>
</tr>
<tr>
<td></td>
<td>3 3.98</td>
<td>3.88</td>
</tr>
<tr>
<td>Temperature (°C)</td>
<td>1 40.08</td>
<td>39.68</td>
</tr>
<tr>
<td></td>
<td>2 41.88</td>
<td>41.92</td>
</tr>
<tr>
<td></td>
<td>3 22.45</td>
<td>23.14</td>
</tr>
<tr>
<td>Exp. RMS Current (A)</td>
<td>1 29.14</td>
<td>28.87</td>
</tr>
<tr>
<td></td>
<td>2 29.52</td>
<td>29.42</td>
</tr>
<tr>
<td></td>
<td>3 20.21</td>
<td>20.23</td>
</tr>
<tr>
<td>Error (%)</td>
<td>1 0.69</td>
<td>1.03</td>
</tr>
<tr>
<td></td>
<td>2 -0.49</td>
<td>-0.02</td>
</tr>
<tr>
<td></td>
<td>3 0.28</td>
<td>-1.00</td>
</tr>
</tbody>
</table>

over time. Thus, temperature control has been demonstrated to reduce capacity mismatch in parallel-connected LFP batteries.

Figure 3.5 shows the standard deviation of the cell capacities $\sigma_{pack}$ versus time. The pack capacities converge as the experiment progresses, resulting in a 48% reduction in $\sigma_{pack}$. 
3.5 Temperature control for simultaneous life maximization and capacity balance

The experimental results confirm theoretical predictions that differential temperature can be used to balance the capacity of parallel-connected cells. Using the validated model, we investigate whether temperature control can simultaneously balance capacity and extend the life of these packs.

Figure 3.6 shows the sensitivity of the current distribution to temperature and
capacity mismatch. As observed in the experiment, the current distribution is much more sensitive to temperature mismatch than capacity mismatch. Thus, relying on the natural self-balancing of the capacity mismatch may not be sufficient to counter unbalancing effects. Temperature difference, however, can significantly accelerate balancing, with 20°C causing almost 30% increase in differential current. Higher temperatures also accelerate aging so cooling the lower capacity cell has the potential to accelerate capacity balancing and extend peak life.

For simplicity, we consider a battery pack with two cells connected in parallel, current distribution transfer functions (Eq. 3.4) simplify to

\[
\frac{I_{\text{cell}_1}(s)}{I_{\text{pack}}(s)} = \frac{G_{\text{cell}_2}(s)}{G_{\text{cell}_1}(s) + G_{\text{cell}_2}(s)}
\]

\[
\frac{I_{\text{cell}_2}(s)}{I_{\text{pack}}(s)} = \frac{G_{\text{cell}_1}(s)}{G_{\text{cell}_1}(s) + G_{\text{cell}_2}(s)}
\]

**Case study parameters:** Four cases are studied to quantify the effect of temperature on capacity mismatch and life. For all four cases one cell start at 4.3Ah and the other at 4.0Ah. End of life (EOL) is defined as when one cell reaches 80% of the fresh cell capacity (4.5Ah). Environmental temperature is 30°C. The four cases:

**Case 1:** Both batteries are operated at the environmental temperature (30°C) until battery pack EOL.

**Case 2:** Both batteries are cooled to operate at room temperature (25°C) until battery pack EOL.

**Case 3:** Cell 1 and 2 are operated at room and environmental temperature, respectively, until both cells have the same capacity. Then, both cells are operated at the environmental temperature until EOL.
Figure 3.7. Simulated battery capacity change in Case 1 (a), Case 2 (b), Case 3 (c), and Case 4 (d). Cell 1 capacity fade (black solid line), Cell 2 capacity fade (blue solid line), Cell 1 temperature (green solid line), and Cell 2 temperature (red dashed line). EOL at 3.6Ah is marked by the magenta dashed line.

Table 3.2. Case Study Results

<table>
<thead>
<tr>
<th>Case #</th>
<th>Pack Life (Years)</th>
<th>Mismatch Reduction (%)</th>
<th>Cooling Energy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case 1</td>
<td>2.09</td>
<td>10</td>
<td>None</td>
</tr>
<tr>
<td>Case 2</td>
<td>4.70</td>
<td>10</td>
<td>Highest</td>
</tr>
<tr>
<td>Case 3</td>
<td>3.52</td>
<td>100</td>
<td>Less than Case 2</td>
</tr>
<tr>
<td>Case 4</td>
<td>4.71</td>
<td>100</td>
<td>Less than Case 2, more than Case 3</td>
</tr>
</tbody>
</table>

**Case 4:** Cell 1 and 2 are operated at room and environmental temperature, respectively, until both cells have the same capacity. Then, both cells are operated at the room temperature until EOL.
In each case, a continuous UDDS cycle is the battery current input. The battery pack performance and degradation model, validated in the experimental study, are used to quantify the capacity mismatch evolution and capacity fade.

Figure 3.7 shows the simulated capacity change of the individual cell versus time for the four cases. In Case 1 and 2, both batteries degrade at a similar rate, but the rate of degradation of the battery pack is higher in case 1. In both cases, capacity mismatch narrows by roughly 10%. This reduction is solely due to the current redistribution caused by the capacity mismatch because both cells have the same temperature. If the temperature is low, the overall capacity fade rate is less, resulting in 4.70 years versus 2.1 years of life for Case 2 and 1, respectively. In Case 3 and 4, the lower capacity battery is cooled at room temperature until both batteries have the same capacity. Cell 1 degrades more slowly and their capacities converge over time. In case 3, both batteries are kept at the higher, environmental temperature after capacity is matched. This increases the degradation rate of the battery pack, reducing pack life compared to Case 4. Table 3.2 summarizes the findings of the case study.

Battery capacity mismatch has less impact on the current distribution and battery capacity fade, so self-balancing is not effective and temperature control is required to balance the battery pack before EOL. In isothermal Cases 1 and 2, the capacity mismatch is only reduced by 10%. Case 4 is the best for maximum battery life and capacity mismatch reduction with moderate energy consumption for battery pack cooling.

### 3.6 Conclusion

The study develops and validates a battery performance and aging model for parallel-connected cells. The model predicts the capacity evolution within 0.60%
over 1500 HEV cycles. The experimental results show that around 18°C temperature difference can reduce the capacity mismatch by 48% over 1500 HEV cycles. Finally, differential cooling of parallel-connected cells can reduce capacity mismatch and extend life.
Chapter 4

Model-based Sizing of Battery Packs for Minimum Cost

4.1 Introduction

Temperature plays a critical role in the performance and aging of Li-ion batteries. High temperatures will accelerate the battery aging [26], but reduce the impedance of the battery pack [29] that enhances the battery performance. Battery aging mechanism is well explained in the literature with different models available to predict battery degradation and SOH, but the literature on selecting the battery capacity and operating temperature is scarce.

Specifically, this study answers three questions for HEV batteries using LFP chemistry.

1. How does one select battery pack size to minimize the cost of operation for a specified battery temperature and usage?

2. How does one select battery temperature to maximize the battery life for a specified battery pack size and usage?
3. How does one select battery pack size and operating temperature for a specified battery life requirement and usage?

4.2 Simulation

A battery pack with 60 LFP cells in series is considered for the simulations. LFP batteries have 3.2 V as nominal voltage so the overall voltage of the battery pack is 192 volt. The capacity of the individual cells ranges from 3.5 Ah to 8.00 Ah for simulations. Thus, simulations are performed for battery packs ranging from 0.672 kWh to 1.536 kWh. The temperature of the battery pack is varied from 25° Celsius to 45° Celsius. The underlying ESPM and aging model have been previously validated for this range so the simulation results are experimentally repeatable. Simulations are performed for 5% usage, 50% usage, and 100% usage. They represent real life scenarios: 5% usage for personal vehicle (1 hr/day), and 50% and 100% usage for different commercial applications. 4280 seconds long charge sustaining HEV cycle Urban Dynamometer Driving Schedule (UDDS) is initialized at 50% SOC for the simulation study [29]. Maximum pulse in the HEV cycle is 70 Amp and minimum pulse is -60 Amp. Simulations are performed under following assumptions:

1. All cells in the battery pack are similar, and the number of cells connected in series is fixed.

2. The EOL of a battery pack is based on two criteria: (1) Remaining capacity or SOH = 80%, (2) voltage greater than 3.6 V, the maximum allowable voltage for LFP chemistry.

3. Battery is at room temperature when not being used.
4. Battery usage is defined as the percentage of the time when UDDS cycle is running, e.g. 50 percent battery usage means the battery is in use for 12 hours per day.

4.3 Results and Discussion

Figure 1.2 shows that the effective impedance of a battery pack reduces with increasing battery temperature. The reduced impedance reduces the maximum voltage during an HEV cycle so the battery is able to live longer. On the other hand, higher temperatures increase the degradation rate. So there is a temperature that will give maximum life.

Battery capacity, operating temperature, life, and usage are correlated and cannot be selected independently. Figures 4.1 and 4.2 explain how to design a battery pack for minimum cost or maximum life. Figure 4.1(a), (b) and (c) show simulation results at 50% battery usage. Figure 4.1(d) shows the possible battery size reduction for different battery usage if the battery is operated at the optimal temperature. Next, we use these simulation results to answer the three questions posed in the introduction.

1. **How does one select battery pack size to minimize the cost of operation for a specified battery temperature and usage?** Figure 4.1(a) shows that as the battery pack capacity increases, battery life increases for a fixed temperature operation. For example, at 25° Celsius, if the battery capacity is less than 4.5 Ah, battery pack life is zero because in the very first HEV cycle the voltage exceeds maximum allowable voltage. As battery pack size increases, battery life increases until battery capacity reaches 5.27 Ah. Above 5.27 Ah, there is no improvement in the battery life for any additional battery capacity. Figure 4.1(b) shows the cost of different capacity batteries at 25° Celsius. The cost is high if
the battery pack capacity is below 5.0 Ah. The cost is minimum when battery pack capacity is 5.27 Ah. Above 5.27 Ah, the cost of the battery pack increases. Figure 4.1(a) also shows there is no reason to have a large battery pack at high temperatures. For example, if the battery operating temperature is 45° Celsius then the life of a 3.5 Ah battery pack is same as the life of an 8.0 Ah battery pack. Higher battery capacity does reduce usage aging because it reduces the C-rate for a fixed input current profile. But at higher temperatures, calendar aging dominates usage aging (Figure 4.3), and increasing the battery capacity will not improve the battery life. Again figure 4.1(b) shows the cost of different capacity batteries at 45° Celsius. It is clear that there exists a minimum cost battery pack for a specific operating temperature and usage, and the cost of deviation from the minimal cost battery pack is higher at a higher temperature.

2. How does one select battery temperature to maximize the battery life for a specified battery pack size and usage? In figure 4.1(a), two limits govern the battery's EOL: Voltage and SOH. At lower temperatures and lower battery capacities, the voltage exceeds EOL limit before SOH reaches 80%. Consider a 4.5 Ah battery pack at 25° Celsius, it exceeds the voltage limit in the very first HEV cycle. If the temperature of this battery pack is 30° Celsius, the battery can cycle. As battery degrades, its capacity reduces, which increases its effective C-rate and internal resistance. The battery eventually exceeds the voltage EOL limit after 4.2 years of cycling at 30° Celsius. At 31.1° Celsius, the 4.5 Ah battery pack would have a maximum possible life of 4.95 years, and both EOL limits (Voltage and SOH) exceed simultaneously. This temperature is the point of maximum life and minimum cost. If operating temperature is higher than this, SOH will reach 80% before voltage exceeds the EOL limit, and battery pack life will reduce. At 35° Celsius, the life of the 4.5 Ah battery pack is 3.06 years.
3. How does one select battery pack size and operating temperature for a specified battery life requirement and usage? Figure 4.1(c) shows the optimal battery temperature ($T_{opt}$) and optimal battery size ($C_{opt}$) for the required battery life. It also shows the battery size required ($C_{rt}$) if the battery operating temperature is room temperature. Serving the warranty period at the minimum cost is desirable in the commercial applications. It is found that room temperature is not always the desired point of operation. Sometimes it is beneficial to use a smaller battery pack at high temperature. For 4 years life requirement, a 4.32 Ah battery pack at 32.8° Celsius or a 4.95 Ah battery pack at room temperature will be sufficient. In this case, a 12.7% battery pack size reduction is possible by increasing the battery temperature. The gap between optimal battery pack size $C_{opt}$ and pack size required $C_{rt}$ for room temperature operation is the reduction in the battery pack size. Figure 4.1(d) shows the possible size reduction for different usage and life requirements. For a 4.5 years life requirement and 5% usage (typical usage for personal vehicles), 30% battery pack reduction is possible if the battery is operated at 45° Celsius. If the life requirement is high, battery should be operated at 25° Celsius as $C_{opt}$ converges to $C_{rt}$, and $T_{opt}$ converges to room temperature.

Simulation is performed from 3.5Ah to 8.0Ah with a step of .01Ah, and temperature range from 25°C to 45°C with step of 0.2°C. Total time taken for the simulations on a quad core computer with 3.4Ghz clock speed and 16Gb RAM was 68 hours.
Figure 4.1. LFP simulation results: (a) Battery pack life at different capacities and at different operating temperatures for 50% battery usage. Each curve represents an equi-capacity plot. (b) Battery pack cost at different temperatures and at different capacities for 50% battery usage. Each curve represents an equi-temperature plot. Cost is calculated in Ah/day, and the cost in $ depends on the battery $/Ah. (c) Figure shows the required minimum battery pack capacity $C_{opt}$ and operating temperature $T_{opt}$ for a specified battery life at 50% battery usage. Figure also shows the battery capacity $C_{rt}$ required if the battery temperature is room temperature. (d) Effect of battery usage on the battery pack size reduction.

4.4 Design Process for Maximum Life or Minimum Cost Pack

Figure 4.2 shows the process of selecting battery capacity and temperature. If there is no active temperature control available then one should determine the
Figure 4.2. Design process to select maximum life and minimum cost battery pack

Determine the expected battery operating temperature and use it with figure 4.1(c) to select the optimal pack size for maximum life and minimum cost. If active temperature control is available, and maximum life is desired, then the battery should be operated at 25°C Celsius. The required battery pack size at 25°C Celsius can be selected using Figure 4.1(c). This design will give the maximum life but keeping battery pack at 25°C Celsius will have addition cost of cooling so the design might not be the most cost efficient. If active temperature control is available, and minimum cost is desired, then cost of cooling of the battery pack must be included. The cost of cooling $C_T$ will depend on cooling design and individual manufacturer. To select the minimum cost pack, one should first calculate the required temperature $T_{req}$ for the overall minimum cost as explained in the flow chart, then use figure 4.1(c) to select the optimal size.
Figure 4.3. Calendar aging contribution in the overall battery aging. Plot is for 100 percent battery usage. Calendar aging contribution in overall aging increases when battery capacity and battery temperature is high for the required temperature $T_{\text{req}}$.

### 4.5 Conclusion

The room temperature is not always desired operating temperature for minimum cost, and suitable operating temperatures depend on the application and life requirements. For low usage and fixed life requirement, it is beneficial to operate a small battery pack at high temperature, but for higher life requirement, the higher battery temperature is detrimental. This study develops a process for selecting the battery size and operating temperature under different conditions. It also shows that calendar aging contribution in the overall aging increases when battery capacity and the temperature is high.
Chapter 5  |  Real Time Temperature Control to Improve Lithium Ion Battery Charge Acceptance

5.1 Introduction

The battery performance and aging depend on the battery operating temperature and usage. High temperatures will accelerate the battery aging but reduce the battery impedance. In this study, two criteria for the battery EOL are considered. First, EOL occurs when the battery SOH reaches 80% of the rated capacity. Second, EOL occurs when the maximum battery voltage exceeds the maximum allowable voltage for LFP chemistry (3.6 V). The BMS does not allow a battery to fully accept the applied current pulse if the battery voltage exceeds 3.6 V. Higher battery temperature reduces the battery internal resistance which reduces the maximum battery voltage, and the battery is able to accept more charge. Thus, by increasing the temperature, the battery charge acceptance and performance can be improved. This study develops real-time temperature control strategies to improve
the battery charge acceptance and reduce the battery pack size. Simulations are performed using experimentally validated models to show that by increasing the battery temperature at higher battery SOC or higher input current, the battery performance can be improved.

### 5.2 Strategies

The battery performance and aging models are combined together to simulate battery life at different battery capacity and temperature. The UDDS cycle is used as the HEV cycle for the simulations, and the simulation results are shown in Figure 5.1. At higher battery temperature, the battery life decreases due to the accelerated aging, but a smaller battery pack is able to provide the required cycle due to the improved performance. Figure 5.1 shows that a 5.5 Ah battery pack
Figure 5.2. Strategy 1 – Higher battery temperature at higher battery SOC. (a) Voltage response. (b) Input current pulse.

has 9.4 years of life at 25° Celsius but only 10 months of life at 45° Celsius. On the other hand, a 4.00 Ah battery pack is not able to provide the required current at 25° and 30° Celsius without exceeding the maximum allowable voltage, but it is able to provide the required current at 40° Celsius. The battery life increases initially as the battery capacity increases at all temperatures, but the battery life
Figure 5.3. Strategy 2 – Higher battery temperature for higher current pulses. (a) Voltage response. (b) Input current pulse. (c) Battery pack temperature.

gets saturated and it does not increase further by increasing the battery capacity. This happens because the battery calendar aging starts to dominate and regardless of the battery pack size, a battery will degrade to 80% SOH in a certain time.

This study develops two real-time temperature control strategies to reduce the
battery pack size and improve performance without affecting the battery life. Both strategies increase the battery temperature judiciously when the battery voltage is more likely to exceed the maximum allowable battery voltage. Increasing the battery temperature will increase the battery charge acceptance.

### 5.2.1 Strategy 1

Figure 5.2 shows the first strategy to improve the battery charge acceptance by increasing the battery temperature at high SOC. Consider a 10C charge pulse for 15 seconds for a 4.5 Ah LFP battery at room temperature. The battery is able to
provide the pulse at 50% SOC, but not at 70% SOC. The maximum battery voltage exceeds 3.6 V at 70% SOC. At higher SOC, the battery open circuit voltage (OCP) is higher, and maximum battery voltage is more likely to exceed the maximum allowable battery voltage. Figure 5.2 also shows that if the battery temperature is increased to 32° Celsius, the battery is able to provide the same pulse at 70% SOC due to the reduced internal resistance at a higher temperature.

5.2.2 Strategy 2

Figure 5.3 shows the second strategy to improve the battery charge acceptance by increasing the battery temperature at high current pulses. Current profiles for the HEV batteries consists of many 4-5 seconds long high and low pulses. The majority of the battery pulses are relatively low, but battery life will be determined by the high pulses due to the voltage based EOL. Consider a pulse shown in Figure 5.3(b) for a 4.5 Ah battery. The battery voltage exceeds 3.6 V if the battery temperature is 25° Celsius during the entire pulse. When the battery temperature is increased by 5° Celsius in the vicinity of the high pulse, the battery voltage does not exceed 3.6 V for the same input, and the battery is able to provide the required pulse.

It is possible to improve the battery charge acceptance by increasing the battery temperature for all time, but it will reduce the battery life substantially as shown in figure 5.1. In these strategies, the temperature is only raised for some time to improve the battery performance and minimize the effect of the temperature on the battery degradation. Improved performance helps to reduce the battery pack size because if the battery pack size is reduced by 10%, battery SOC swing and effective battery C rate will increase by 10% for the same input profile.
5.3 Simulation

Simulations are performed for the battery packs of different capacities at different temperature and usage. Battery usage is defined as percentage of time the battery is in use, e.g. 50% battery usage means the battery is operated 12 hours per day. Battery simulation is performed for 5% usage (1 hour per day, typical for passenger car) and 50% and 100% usage representing commercial vehicles. Batteries are simulated under following assumptions:

1. Battery current profile is known beforehand. UDDS profile is used for the simulations.

2. The battery temperature control mechanism is available, and the time constant to heat up and cool down the battery pack is 5 seconds. It is also assumed that a battery reaches the required temperature in 4 time constants.

3. The battery is at room temperature when the high temperature is not required.

4. The maximum allowable battery temperature is 45° Celsius due to safety concerns.

5. EOL condition 1 – Maximum battery voltage > 3.6 V at the maximum allowable battery temperature.

6. EOL condition 2 – SOH < 80%.

Two strategies developed in the last section are combined for the simulations. The battery temperature is raised whenever the battery voltage exceeds 3.6 V either due to high pulse or high SOC. Figure 5.4 shows the flow chart of the simulation. Initially, a battery is assumed to be at room temperature, and the battery performance model is used to get the battery voltage during the cycle. If
Figure 5.5. (a) Battery temperature profile with the battery heat up and cool down time constant of 5 seconds. (b) Battery input current. (c) Battery temperature profile with the battery heat up and cool down time constant of 25 seconds.
the battery voltage is below 3.6 V, the battery is cycled for 100 cycles and its SOH is updated. Cycling will increase the battery internal resistance and the battery voltage is likely to eventually exceed 3.6 Volt. The performance model is again used to get the battery voltage during the cycle. If the battery voltage is above 3.6 V, the battery temperature is raised by 1°C where the battery voltage exceeds 3.6 V, and the performance model is again used with the new temperature profile. Figure 5.5(a) shows the battery temperature during the first 500 seconds of the 4200 seconds long UDDS profile for a 3.82 Ah battery. The battery temperature
is raised at three times so that the battery will be able to serve the UDDS cycle. Simulations are stopped when the battery SOH reached 80% or the battery voltage exceeds 3.6 Volt at the maximum allowable battery temperature.

5.4 Results and Discussion

Figure 5.5(a) shows the battery temperature of a 3.82 Ah battery at different SOH for the first 500 seconds of UDDS cycle. There are three high pulses in first 500 seconds that cause the battery voltage to exceed 3.6 Volt in the very first cycle. Therefore, the battery will not be able to provide any cycle without increasing the battery temperature. By increasing the battery temperature during those high pulses, the battery voltage gets reduced, and the battery can provide some HEV cycles. The simulations show that at least 4.77 Ah battery pack is required to provide the UDDS cycle at room temperature operation. Hence, in this case, battery size is reduced from 4.77 Ah to 3.82 Ah by increasing the battery temperature. Figure 5.5(a) also shows that the battery temperature needs to be increased further as the battery SOH decreases. As the battery degrades, its internal resistance increases which in turn increases the maximum battery voltage. Therefore, by further increasing the battery temperature, maximum battery voltages are kept within the limit.

Increasing the battery temperature reduces the battery life based on SOH. Therefore, simulation results are presented as how much battery pack size can be reduced by using these strategies for a fixed battery life. In the simulation, $C_{rt}$ is defined as the required battery pack size if the battery is operated at the room temperature, and $C_{opt}$ is defined as the minimum battery pack size required if the battery temperature is actively controlled. Figure 5.6(a), (b), and (c) show $C_{rt}$ and $C_{opt}$, and Figure 5.6(d) shows the battery pack size reduction for different battery
usage and life. There is a trade-off between battery size reduction and battery life requirement. The main results of the simulations are as follows:

1. At low battery usage (5% usage), 30% battery pack size reduction is possible for almost all the battery life requirements.

2. At high battery usage (50% and 100%), 30% battery pack size reduction is possible if the life requirement is 5 years or less.

3. At high battery usage and high life requirement, less size reduction is possible because high temperature starts to affect the battery life.

4. No battery pack size reduction is possible if the maximum battery life is desired because $C_{rt}$ and $C_{opt}$ converge for the maximum battery life.

5. Increasing the battery pack size increases the battery life to an extent. After a limit, battery life can not be increased by increasing the battery size as calendar aging and SOH based EOL start to dominate.

Simulation results discussed so far assume that the time constant to heat up and cool down the battery pack is 5 seconds. Figure 5.5 also compares the effect of the time constant on the battery temperature. Two simulations are performed using the same method as explain in the flow chart except for different time constants. The time constant is 5 seconds and 25 seconds in the simulation 1 and 2 respectively. If the time constant is large, the battery will tend to be at higher temperature for more time. Figure 5.5 shows that there are two large pulses within 150 seconds. If the time constant is small, the battery is at room temperature between those pulses, but if the time constant is large, the battery will be at a higher temperature between those two pulses (Figure 5.5(c)). Higher battery temperature will affect the battery life, and the battery will degrade faster. Figure 5.7 compares the relative

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Figure 5.7. Effect of the battery heat up and cool down time constant on the battery pack size reduction. Simulation 1 and 2 have time constants of 5 seconds and 25 seconds respectively.

reduction in the battery pack size in both simulations for 50% battery usage. In simulation 2, the battery is at high temperature for more time and degrades faster; therefore, a relatively low battery pack size reduction can be achieved using the strategies developed in this study. Thus, the time constant to heat up and cool down the battery plays an important role. If the time constant is too high, then there would not be any improvement due to these strategies. Researchers have developed the batteries to heat up the battery pack very fast, almost 1°C per second [37].
5.5 Conclusion

The room temperature is not always a desired operating temperature for the minimum battery size and cost. By actively controlling the battery temperature, simulation results predict battery pack size reduction of up to 30%. The battery pack size reduction also depends on the battery usage and life requirements. This study develops a process to select the battery pack size and the corresponding battery temperature profile based on the battery current profile for different applications.

The study explores that increasing and decreasing the temperature at a faster rate will improve the battery performance, extend life and reduce pack size. Results can motivate the innovative battery designs which enable the fast cooling and heating of the battery pack.
Chapter 6

Battery

Safety: Nail Penetration And Overcharge Tests

6.1 Introduction

Higher energy density in lithium ion batteries brings major safety concerns. Safety is a key factor in achieving widespread acceptability of lithium ion battery powered electrified vehicles. Internal short circuits, external short circuits, overcharging, and over-heating are the primary battery abuse conditions that pose safety concerns.

This study presents an iNail design to conduct battery nail penetration tests. The iNail can capture the highest temperature measurement at the point of penetration where the nail penetrates through the cell. Highest temperature affects the thermal runaway in the battery. The iNail can also be used for pack level nail penetration tests where pack thickness is considerably large and a single thermocouple at the nail tip (as proposed by Hatchard et al. [68]) is not sufficient to capture temperature readings at different locations of the pack.
This study also explores the battery overcharge for NCA and NCM chemistry cylindrical cells. The objective is to analyze the effect of battery chemistry, C-rate, temperature, and aging on the battery overcharge.

6.2 Intelligent Nail Design for Lithium Ion Battery

Nail Penetration Test

The iNail (shown schematically in Figure 6.1) measures temperatures during nail penetration test in a more convenient and efficient manner. The iNail can be conveniently disassembled into three parts where multiple sensors (thermocouples, strain gauges, etc.) can be conveniently placed for reliable and efficient data collection. Mounting multiple thermocouples inside the iNail avoids precise position control of the nail during the test and provides temperature readings along the
cross-section of the cell. A prototype stainless steel iNail is manufactured and presented. Multiple thermocouples are placed inside the iNail and on the cell surface. Nail penetration tests are conducted on fully charged 4 Ah gr/NCM pouch cells. The iNail successfully recorded the temperature time history around the penetration point during the tests. Results are reported in terms of time histories of temperature and voltage during the nail penetration tests. Successful pack level nail penetration tests (three pouch cells in parallel) have also been performed with the newly designed iNail.

Figure 6.2 shows the 3D drawings of the disassembled and assembled iNail. The iNail consists of three sections: top, middle, and bottom and the nail tip. The top section goes into the drill chuck and holds the complete nail. The bottom part of the top section has external threads to fit with the iNail middle section. The middle section contains three openings for sensor wiring along with two internally threaded ends to fit with the top and bottom sections. The bottom section is specially designed and can be split into two parts (see cross-sectional view of the bottom section in Figure 6.2). This design allows easy placement of different sensors like thermocouple, strain gauge, etc. The two ends of the bottom section are also externally threaded to be assembled with the iNail tip and the middle section. Figure 6.2 also shows the complete assembled iNail. The geometric design parameters of the iNail are given in Table 6.1.

6.3 Nail Penetration Test: Experimental Setup

Figure 6.3(c) shows the complete assembled iNail prototype along with three K-type thermocouples. The iNail is made of stainless steel. Figure 6.3(a) shows the two split parts of the iNail bottom section where four tapered pins are used to keep the two sections together. Figure 6.3(b) shows the installed thermocouples on the iNail
Figure 6.2. Three dimensional drawing of the iNail showing different parts (left) and complete nail (right).

bottom section. Figure 6.3(d) shows the iNail is clamped with the drill chuck of the nail penetration tester. Figure 6.3(e) shows a nail penetration test.

For this test, four thermocouples equally spaced 20 mm apart are placed along the center line of a fully charged 4 Ah gr/NCM pouch cell. There is no safety device attached to the cell internally or externally. Three thermocouples spaced 25 mm apart are placed longitudinally inside the bottom section of the iNail (see Figure 6.3(b)). Cell voltage is also measured during the nail penetration test. The
Figure 6.3. Experimental setup: (a) iNail bottom section, (b) iNail bottom section with thermocouples installed, (c) complete iNail prototype, (d) single cell penetration test setup, and (e) three cell parallel pack nail penetration test.

<table>
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<th>Parameter</th>
<th>Value</th>
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</tr>
<tr>
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</tr>
</tbody>
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penetration speed was 10 mm/s.

6.4 Nail Penetration Test: Results and Discussion

Figure 6.4(a) shows the location of the thermocouples inside the iNail and on the test cell. Figure 6.4(b) is showing the temperature time histories along the longitudinal direction of the iNail bottom section during the penetration test. The temperature at the point of penetration ($T_5$) rises quickly to $92^\circ C$, immediately after the penetration primarily due to a large amount of ohmic heat generation. The temperature then reduces to $85^\circ C$ and rises up to a maximum of $107^\circ C$ before
falling down. This temperature fluctuation shows the cell dynamics during the nail penetration and involves repeating heating and cooling processes. Temperatures away from the point of penetration $T_6$ and $T_7$ follow the same trend, but with a lower peak due to the more exposed area around the nail which facilitate heat transfer. Figure 6.4(c) shows the time history of the temperature of the cell which shows a monotonous increase in temperature up to a peak around $80^\circ C$ and then decrease. Figure 6.4(d) plots the maximum temperatures ($T_1$ and $T_5$) and the cell voltage during the nail penetration test. Nail temperature is higher compared to the cell temperature due to high ohmic heat generation at the small nail shorting area and less heat dissipation from the shorting area. The rate of temperature increase (temperature response) is also significantly higher at the nail compared to the cell. The immediate drop of cell voltage after the penetration confirms the internal short following the penetration.

### 6.5 Overcharge Test: Experimental Set Up

Battery overcharge test of the cylindrical cells for 2 different chemistries was performed under different environmental and operational conditions. Fresh and aged cells were overcharged for NCA and NCM chemistry at 1C and 10C charge rate at $45^\circ C$ and $-20^\circ C$. Cells were first charged up to 100% SOC at room temperature using CC-CV charging method. Then their temperature raised to $45^\circ C$ or decreased to $-20^\circ C$ as required, and maintained there. Overcharge was done at constant current rate 1C or 10 C as required till any failure occurs. Failure was defined as fire, smoke or cell is not able to take any more current. As tests require higher and lower temperature operation, heater and dry ice were used for heating and cooling. All cells were 18650 cylindrical cell. For NCA chemistry the capacity was 2.9 Ah and for NCM chemistry the capacity was 2 Ah.
Figure 6.5. Battery overcharge test setup (a) Set up for overcharging and nail penetration, (b) Heater used to raise Cell temperature, and (c) Dry ice used to reduce the Cell temperature.

Figure 6.5 shows the experimental set up for overcharge tests. The experiments were conducted in the open space as overcharge tests are safety hazard. Cells were placed in the air press fitted with a nail to penetrate the cell. After overcharge, a nail penetration setup is used to penetrate the cell for disposal. During the experiment, battery internal safety mechanism got triggered at it is not possible to discharge it normally so nail penetration is performed to discharge the batteries.
6.6 Overcharge Test: Results and Discussion

Figure 6.6 - Figure 6.17 shows the battery overcharge experiments at different conditions. Table 6.2 and 6.3 summarizes the maximum voltage and maximum SOC in each experiment. Few observations for the overcharge test are as follows:

1. All batteries were commercial cylindrical cells which have current interrupter deceive (CID) as a safety device. In all experiments, CID operated before any visual failure.

2. Amount of the current accepted before CID activated depends on the C rate. At low C rate, the battery is able to absorb more charge, thus lower C rate is relatively safer for battery overcharge.

3. At high C rate (10C) the temperature rise is very sharp and CID operated very soon (within 5 percent of SOC rise).

4. Voltage rise depends on the C rate and operating temperature. Initial voltage rise at -20°C was very sharp for both 1C and 10 C because of high internal resistance.

5. Experiments show that aged cells are able to absorb more charge before CID is activated.
Table 6.2. NCA vs NCM overcharge at different conditions

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<thead>
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<th>Operating Condition</th>
<th>NCA (Fresh)</th>
<th>NCM (Fresh)</th>
</tr>
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<tbody>
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<td></td>
<td>Max V</td>
<td>Max SOC (%)</td>
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<tr>
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<tr>
<td>Temperature = 45°C</td>
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<td>115.17</td>
</tr>
<tr>
<td>Rate = 10C</td>
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<td>Temperature = 45°C</td>
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</tr>
<tr>
<td>Rate = 1C</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Temperature = -20°C</td>
<td>5.90</td>
<td>104.15</td>
</tr>
<tr>
<td>Rate = 10C</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Temperature = -20°C</td>
<td>9.55</td>
<td>101.67</td>
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</tbody>
</table>
Table 6.3. NCM (Fresh) vs NCM (Aged) overcharge at different conditions

<table>
<thead>
<tr>
<th>Operating Condition</th>
<th>NCM (Aged)</th>
<th>NCM (Fresh)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Max V</td>
<td>Max SOC (%)</td>
</tr>
<tr>
<td>Rate = 1C Temperature = 45°C</td>
<td>5.05</td>
<td>167.83</td>
</tr>
<tr>
<td>Rate = 10C Temperature = 45°C</td>
<td>5.12</td>
<td>155.25</td>
</tr>
<tr>
<td>Rate = 1C Temperature = -20°C</td>
<td>5.24</td>
<td>213.37</td>
</tr>
<tr>
<td>Rate = 10C Temperature = -20°C</td>
<td>5.85</td>
<td>158.28</td>
</tr>
</tbody>
</table>
Figure 6.6. Overcharge Test 1: NCA chemistry fresh cell with 2.9Ah capacity overcharged at 1C rate with 45°C initial cell surface temperature. Maximum battery voltage is 5.23V. CID operated after 546 seconds at 115.17% battery SOC. Cell surface temperature (dashed magenta and dashed-dotted red), environmental temperature (dotted green), battery terminal voltage (blue dotted), and battery current (solid black).
Figure 6.7. Overcharge Test 2: NCA chemistry fresh cell with 2.9Ah capacity overcharged at 1C rate with -20°C initial cell surface temperature. Maximum battery voltage is 5.90V. CID operated after 150 seconds at 104.15% battery SOC. Cell surface temperature (dashed magenta and dashed-dotted red), environmental temperature (dotted green), battery terminal voltage (blue dotted), and battery current (solid black).
Figure 6.8. Overcharge Test 3: NCA chemistry fresh cell with 2.9Ah capacity overcharged at 10C rate with 45°C initial cell surface temperature. Maximum battery voltage is 5.73V. CID operated after only 6 seconds at 101.67% battery SOC. Cell surface temperature (dashed magenta and dashed-dotted red), environmental temperature (dotted green), battery terminal voltage (blue dotted), and battery current (solid black).
Figure 6.9. Overcharge Test 4: NCA chemistry fresh cell with 2.9Ah capacity overcharged at 10C rate with -20°C initial cell surface temperature. Maximum battery voltage is 9.55V. CID operated after 6 seconds at 101.67% battery SOC. Cell surface temperature (dashed magenta and dashed-dotted red), environmental temperature (dotted green), battery terminal voltage (blue dotted), and battery current (solid black).
Figure 6.10. Overcharge Test 5: NCM chemistry fresh cell with 2.0Ah capacity overcharged at 1C rate with 45°C initial cell surface temperature. Maximum battery voltage is 5.08V. CID operated after 1859 seconds at 151.64% battery SOC. Cell surface temperature (dashed magenta and dashed-dotted red), environmental temperature (dotted green), battery terminal voltage (blue dotted), and battery current (solid black).
Figure 6.11. Overcharge Test 6: NCM chemistry fresh cell with 2.0Ah capacity overcharged at 1C rate with -20°C initial cell surface temperature. Maximum battery voltage is 5.19V. CID operated after 1888 seconds at 152.44% battery SOC. Cell surface temperature (dashed magenta and dashed-dotted red), environmental temperature (dotted green), battery terminal voltage (blue dotted), and battery current (solid black).
Figure 6.12. Overcharge Test 7: NCM chemistry fresh cell with 2.0Ah capacity overcharged at 10C rate with 45°C initial cell surface temperature. Maximum battery voltage is 5.23V. CID operated after 16 seconds at 104.45% battery SOC. Cell surface temperature (dashed magenta and dashed-dotted red), environmental temperature (dotted green), battery terminal voltage (blue dotted), and battery current (solid black).
Figure 6.13. Overcharge Test 8: NCM chemistry fresh cell with 2.0Ah capacity overcharged at 10C rate with -20°C initial cell surface temperature. Maximum battery voltage is 6.37V. CID operated after 18 seconds at 105.00% battery SOC. Cell surface temperature (dashed magenta and dashed-dotted red), environmental temperature (dotted green), battery terminal voltage (blue dotted), and battery current (solid black).
Figure 6.14. Overcharge Test 9: NCM chemistry aged cell overcharged at 1C rate with 45\(^\circ\)C initial cell surface temperature. Maximum battery voltage is 5.05V. CID operated after 2442 seconds at 167.83% battery SOC. Cell surface temperature (dashed green and dashed-dotted red), environmental temperature (dotted magenta), battery terminal voltage (solid black), and battery current (dotted blue).
Figure 6.15. Overcharge Test 10: NCM chemistry aged cell overcharged at 1C rate with -20°C initial cell surface temperature. Maximum battery voltage is 5.24V. CID operated after 4081 seconds at 213.37% battery SOC. Cell surface temperature (dashed green and dashed-dotted red), environmental temperature (dotted magenta), battery terminal voltage (solid black), and battery current (dotted blue).
Figure 6.16. Overcharge Test 11: NCM chemistry aged cell overcharged at 10C rate with 45°C initial cell surface temperature. Maximum battery voltage is 5.12V. CID operated after 198.9 seconds at 155.25% battery SOC. Cell surface temperature (dashed green and dashed-dotted red), environmental temperature (dotted magenta), battery terminal voltage (solid black), and battery current (dotted blue).
Figure 6.17. Overcharge Test 12: NCM chemistry aged cell overcharged at 10C rate with -20°C initial cell surface temperature. Maximum battery voltage is 5.85V. CID operated after 210 seconds at 158.28% battery SOC. Cell surface temperature (dashed green and dashed-dotted red), environmental temperature (dotted magenta), battery terminal voltage (solid black), and battery current (dotted blue).
Chapter 7  |  Conclusions And Future Work

7.1 Conclusions

The dissertation documents a comprehensive study of the effect of the battery temperature on its performance and aging. This study develops the methods to select the battery pack under different conditions and to control the battery temperature to increase the battery life or reduce the capacity mismatch.

Previously developed ESPM and SEI layer based degradation models are used to study the effect of temperature on different aspects of the battery performance and aging. The models are developed and validated further for different temperature and capacity. In chapter 2, a counter-intuitive hypothesis that increasing the battery temperature can extend the battery life is validated theoretically and experimentally. Two 1°C temperature increases extend the life of a 4.5 Ah LFP cell by 2000 charge sustaining HEV cycles. These small temperature increases allowed the cell to cycle continuously for an additional 3 months. In chapter 3, the models are extended for battery packs. Current distribution among cells connected in parallel is validated at different temperature for different capacity packs. A case study is done to show that temperature control can reduce battery pack mismatch,
and room temperature is not always the best choice for all the considerations.

Experimental results of chapter 2 and 3 extensively validate the models. The performance model is now able to predict the change in the battery output voltage due to capacity fade and temperature change. It is also extended to calculate the current distribution for different capacity packs at different temperatures. The degradation model is validated for a wider range of temperature and state of health. Comprehensively validated models are used for the design of battery capacity and operating temperature. The room temperature is not always a desired operating temperature for the minimum battery size and cost. In chapter 4, it is shown that optimal operating temperatures depend on the application and life requirements. For low usage and fixed life requirement, it is beneficial to operate a small battery pack at high temperature, but for higher life requirement, the higher battery temperature is detrimental. A process is developed to select the battery pack size and operating temperature as per the conditions and requirements. The effect of calendar aging in the overall aging is discussed and at a higher temperature, most of the aging comes from calendar aging. In chapter 5, a temperature trajectory is developed if the current is known beforehand. By actively controlling the battery temperature, battery pack size can be reduced up to 30%. The battery pack size reduction depends on the battery usage and life requirements, for a lower life requirement larger size reduction is possible.

The increase in battery life and performance discussed in this dissertation applies to the HEV applications. It may not be true for EVs due to different requirements for EVs. HEV requires high power for a small amount of time, but EV needs low C-rate current for a longer time. Typical C-rate for HEVs can go as high as 15-20C while for EVs it is less than C/3. Improvement due to battery heating also depends on the battery chemistry. If the battery pack resistance is more sensitive to the
temperature and battery degradation is less sensitive to the temperature, then the strategies developed will be even more effective.

Finally in chapter 6, battery nail penetration and overcharge tests are performed to explore the battery safety. A new nail design is presented and manufactured which can successfully capture the temperature time history at the penetration point during the nail penetration test. Unlike conventional solid nails, the newly designed nail allows performing pack level nail penetration tests involving multiple cells. The Nail design is flexible and requires less effort to install the sensors. Battery overcharge tests show the effect of the battery temperature, chemistry, and C-rate on the battery overcharge. At high C rate, overcharge is more detrimental for both NCM and NCA chemistry, while at low C rate, NCM is more tolerant to overcharge compare to NCA.

7.2 Future Work

The present work extends and validates the aging and performance models from 25°C to 45°C. At lower temperature, other degradation mechanisms become substantial such as lithium plating. Extending the aging and performance models for temperature below room temperature could be done in future. As battery temperature affect the battery life, and different temperature control mechanism can be used to meet different goals, similarly battery SOC set point also affects the battery life. Battery SOC set point can be selected to minimize the battery degradation. It is also possible to reduce battery capacity mismatch by operating different batteries in a battery pack at different SOC. Other possible future work can include battery internal and external short circuit modeling.
Appendix

ESPM Model Equations

Table A.1. Fundamental Electrochemical Governing Equations [1]

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Equation</th>
<th>Boundary conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td>Solid phase $Li^+$ conservation</td>
<td>$\frac{\partial c_s}{\partial t} = D_s \frac{\partial}{\partial r} \left( r^2 \frac{\partial c_s}{\partial r} \right)$</td>
<td>$\frac{\partial \phi_s}{\partial x} \bigg</td>
</tr>
<tr>
<td>Solid phase charge conservation</td>
<td>$\frac{\partial}{\partial x} \left( \sigma_{eff} \frac{\partial}{\partial x} \phi_s \right) = j_{Li} - \sigma_{eff} \frac{\partial \phi_s}{\partial x} \bigg</td>
<td><em>{x=0} = \sigma</em>{eff} \frac{\partial \phi_s}{\partial x} \bigg</td>
</tr>
<tr>
<td>Electrolyte phase $Li^+$ conservation</td>
<td>$\frac{\partial (\epsilon_e c_e)}{\partial t} = \frac{\partial}{\partial x} \left( D^{eff} \frac{\partial}{\partial x} c_e \right) + 1 - \frac{\rho_0}{F} j_{Li} \frac{\partial c_e}{\partial x} \bigg</td>
<td>_{x=0} = \frac{\partial c_e}{\partial x} \bigg</td>
</tr>
<tr>
<td>Electrolyte phase charge conservation</td>
<td>$\frac{\partial}{\partial x} \left( k^{eff} \frac{\partial}{\partial x} \phi_e \right) + \frac{\partial}{\partial x} \left( k^{eff} \frac{\partial}{\partial x} \ln c_e \right) + j_{Li} = 0 \frac{\partial \phi_e}{\partial x} \bigg</td>
<td>_{x=0} = \frac{\partial \phi_e}{\partial x} \bigg</td>
</tr>
<tr>
<td>Butler Volmer</td>
<td>$j_{Li} = a_s i_0 \left[ \exp \left( \frac{\alpha_s F}{RT} \eta \right) - \exp \left( -\frac{\alpha_s F}{RT} \eta \right) \right]$</td>
<td></td>
</tr>
<tr>
<td>Overpotential</td>
<td>$\eta = \phi_s - \phi_e - U$ (6)</td>
<td></td>
</tr>
<tr>
<td>Voltage Output</td>
<td>$V(t) = \phi_s(L, t) - \phi_e(0, t) - \frac{R_c}{A} I(t)$ (7)</td>
<td></td>
</tr>
</tbody>
</table>
### Table A.2. Reduced order ESPM equations [1]

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Transfer function</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Solid phase</strong>&lt;br&gt;Li$^+$ conservation</td>
<td></td>
</tr>
<tr>
<td><strong>Positive particle</strong>&lt;br&gt;$\tilde{C}_{p,e}^p(s) = \frac{21(\frac{s^2}{a_p^p F A_p R_p^p L_p} + \frac{60D_p^p s}{[R_p^p]^{3/2}} + \frac{495[D_p^p]^{10/3}}{[R_p^p]^{5}})}{s^3 + \frac{189D_p^p s^{2/3}}{[R_p^p]^{2/3}} + \frac{3465[D_p^p]^{10/3}}{[R_p^p]^{5}}}$ (8a)</td>
<td></td>
</tr>
<tr>
<td><strong>Negative particle</strong>&lt;br&gt;$\tilde{C}_{n,e}^n(s) = \frac{-21(\frac{s^2}{a_n^p F A_n R_n^p L_n} + \frac{60D_n^p s}{[R_n^p]^{3/2}} + \frac{495[D_n^p]^{10/3}}{[R_n^p]^{5}})}{s^3 + \frac{189D_n^p s^{2/3}}{[R_n^p]^{2/3}} + \frac{3465[D_n^p]^{10/3}}{[R_n^p]^{5}}}$ (8b)</td>
<td></td>
</tr>
<tr>
<td><strong>Solid phase charge conservation</strong></td>
<td></td>
</tr>
<tr>
<td><strong>Positive particle</strong>&lt;br&gt;$J_{p,i}^p(s) = \frac{1}{A_p L_p}$ (9a)</td>
<td></td>
</tr>
<tr>
<td><strong>Negative particle</strong>&lt;br&gt;$J_{n,i}^n(s) = \frac{1}{A_n L_n}$ (9b)</td>
<td></td>
</tr>
<tr>
<td><strong>Electrolyte phase Li$^+$ and charge conservation</strong></td>
<td></td>
</tr>
<tr>
<td>$\frac{\Delta \phi_e(L,s)}{I(s)} = \frac{R_2 s^2 + R_1 s + R_0}{L_2 s^2 + L_1 s + L_0}$ (10)</td>
<td></td>
</tr>
<tr>
<td><strong>Butler Volmer Equation</strong></td>
<td></td>
</tr>
<tr>
<td><strong>Positive particle</strong>&lt;br&gt;$\eta_p(s) = \frac{R_c^p}{a_p^p} \frac{1}{A_p L_p} ; \quad R_c^p = \frac{RT}{F(\alpha_c + \alpha_a) i_0} ; \quad a_p^p = \frac{3\varepsilon_p^p}{R_c^p}$ (11a)</td>
<td></td>
</tr>
<tr>
<td><strong>Negative particle</strong>&lt;br&gt;$\eta_n(s) = \frac{R_c^n}{a_n^p} \frac{1}{A_n L_n} ; \quad R_c^n = \frac{RT}{F(\alpha_a + \alpha_c) i_0} ; \quad a_n^p = \frac{3\varepsilon_n^p}{R_c^n}$ (11b)</td>
<td></td>
</tr>
<tr>
<td><strong>Voltage Output Equation</strong></td>
<td></td>
</tr>
<tr>
<td>$\frac{\tilde{V}(s)}{I(s)} = \frac{\eta_p(s)}{I(s)} + \frac{\partial U}{\partial \tilde{C}<em>{p,e}^p(s)} \frac{\tilde{C}</em>{p,e}^p(s)}{I(s)} - \frac{\eta_n(s)}{I(s)} - \frac{\partial U}{\partial \tilde{C}<em>{n,e}^p(s)} \frac{\tilde{C}</em>{n,e}^p(s)}{I(s)} + \frac{\Delta \phi_e(L,s)}{I(s)} - R_c$ (12)</td>
<td></td>
</tr>
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### Table A.3. Model parameters of a 4.5 Ah LFP cell

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Neg. electrode</th>
<th>Separator</th>
<th>Pos. electrode</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thickness, $L$ (cm)</td>
<td>$3.40 \times 10^{-3}$</td>
<td>$2.50 \times 10^{-3}$</td>
<td>$7.00 \times 10^{-3}$</td>
</tr>
<tr>
<td>Particle radius, $R_s$ (cm)</td>
<td>$2.90 \times 10^{-4}$</td>
<td></td>
<td>$3.65 \times 10^{-6}$</td>
</tr>
<tr>
<td>Active material volume fraction, $\varepsilon_s$</td>
<td>0.55</td>
<td></td>
<td>0.41</td>
</tr>
<tr>
<td>Porosity (electrolyte phase volume fraction), $\varepsilon_e$</td>
<td>0.33</td>
<td>0.40</td>
<td>0.33</td>
</tr>
<tr>
<td>Maximum solid phase concentration, $c_{s,max}$ (mol cm$^{-3}$)</td>
<td>$31.07 \times 10^{-3}$</td>
<td></td>
<td>$22.80 \times 10^{-3}$</td>
</tr>
<tr>
<td>Stoichiometry at 0% SOC, $x_{0%}, y_{0%}$</td>
<td>0</td>
<td></td>
<td>0.76</td>
</tr>
<tr>
<td>Stoichiometry at 100% SOC, $x_{100%}, y_{100%}$</td>
<td>0.80</td>
<td>0.03</td>
<td></td>
</tr>
<tr>
<td>Average electrolyte concentration, $c_{e,avg}$ (mol cm$^{-3}$)</td>
<td></td>
<td></td>
<td>$1.2 \times 10^{-3}$</td>
</tr>
<tr>
<td>Exchange current density, $i_{0,ref}$ (A cm$^{-2}$)</td>
<td>$1.85 \times 10^{-4}$</td>
<td></td>
<td>$8.18 \times 10^{-5}$</td>
</tr>
<tr>
<td>Activation energy of $i_0$ (kJ mol$^{-1}$)</td>
<td>40</td>
<td></td>
<td>25</td>
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<tr>
<td>Charge transfer coefficient, $\alpha_a, \alpha_c$</td>
<td>0.5, 0.5</td>
<td></td>
<td>0.5, 0.5</td>
</tr>
<tr>
<td>$Li^+$ transference number, $t_0^+$</td>
<td></td>
<td></td>
<td>0.38</td>
</tr>
<tr>
<td>Solid phase Li diffusion coefficient, $D_{s,ref}$ (cm$^2$ s$^{-1}$)</td>
<td>$5.29 \times 10^{-11}$</td>
<td></td>
<td>$1.18 \times 10^{-14}$</td>
</tr>
<tr>
<td>Activation energy of $D_s$ (kJ mol$^{-1}$)</td>
<td>4</td>
<td></td>
<td>20</td>
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<tr>
<td>Area, $A$ (cm$^2$)</td>
<td>3580</td>
<td></td>
<td>3487</td>
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</table>
Table A.4. Governing equations of the SEI growth model [2]

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Equation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volumetric current density</td>
<td>$j_n^{Li} = j_n^I + j_n^{sei}$</td>
</tr>
<tr>
<td>Tafel equation (SEI reaction):</td>
<td>$j_n^{sei} = -a_n^s i_{0,sei} \exp \left(-\frac{\alpha_n F}{RT} \eta_{sei}\right)$</td>
</tr>
<tr>
<td>Side reaction overpotential:</td>
<td>$\eta_{sei} = \phi^n_s - \phi_e - U_{sei}^{ref} - \frac{j_n^{Li}}{a_n} R_{sei}$</td>
</tr>
<tr>
<td>B-V reaction:</td>
<td>$\eta_n = \frac{RT}{\alpha_n F} \sinh^{-1} \left( \frac{j_n^I}{2a_n i_{0,n}} \right)$</td>
</tr>
<tr>
<td>Intercalation reaction overpotential:</td>
<td>$\eta_n = \phi^n_s - \phi_e - U_n \left( c^n_{s,e} \right) - \frac{j_n^{Li}}{a_n} R_{sei}$</td>
</tr>
<tr>
<td>Capacity loss:</td>
<td>$\frac{\partial Q_{Loss}}{\partial t} = - \int_0^{L_n} j_n^{sei} A_n$</td>
</tr>
</tbody>
</table>

Table A.5. Aging parameters of a 4.5 Ah LFP cell

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
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</thead>
<tbody>
<tr>
<td>Side reaction equilibrium potential, $U_{sei}^{ref}$ (V)</td>
<td>0.4</td>
</tr>
<tr>
<td>Side reaction exchange current density, $i_{0,sei}$ (A cm$^{-2}$)</td>
<td>$7.19 \times 10^{-11}$</td>
</tr>
<tr>
<td>SEI layer Molar mass, $M_{sei}$ (kg mol$^{-1}$)</td>
<td>0.162</td>
</tr>
<tr>
<td>SEI layer density, $\rho_{sei}$ (kg cm$^{-3}$)</td>
<td>$1690 \times 10^{-6}$</td>
</tr>
<tr>
<td>SEI ionic conductivity, $\kappa_{sei}$ (S cm$^{-1}$)</td>
<td>0.0575</td>
</tr>
<tr>
<td>Side reaction activation energy, $E_{a,sei}$ (J mol$^{-1}$)</td>
<td>$1.38 \times 10^5$</td>
</tr>
</tbody>
</table>
Bibliography


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August 2010 - July 2011 Executive Trainee
NTPC Ltd., India
May 2009 - July 2009 Summer Internship
FEV India Pvt Ltd., India