Optimal Design of Li-Ion Batteries through Multi-Physics Modeling and Multi-Objective Optimization

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Battery design variable optimization can significantly affect battery capacity, discharge specific power, and discharge specific energy. However, many design variables need to be taken into consideration, which requires intensive computation and simulation. Our previously developed comprehensive battery degradation model is utilized in this optimization study via parallel computing. A three-electrode cell is developed for model validation over long term cycling. The objectives of optimization are maximizing discharge specific power and specific energy as well as minimizing capacity loss. Several design variables (e.g., thickness, particle size, and porosity) are optimized through a modified Elitist Non-Dominated Sorting Genetic Algorithm (NSGA-II). The obtained Pareto-optimal solutions that show electrode thickness, particle sizes, porosity, and conductivity are the battery design variables that can significantly affect battery performance. In addition, a sensitivity analysis suggests that a thicker electrode and a smaller particle size can improve battery performance. The optimized batteries have a better performance over 750 cycle’s simulation: less SOC swing and less reduction of capacity. The design optimization framework developed herein can be modified and applied to various type of batteries with different optimization objectives and battery design variables.

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More than one million electric vehicles (EVs) have been sold worldwide in the last two decades.1 The trend of vehicle electrification is likely to continue in the future with battery technology innovation. The increasing number of EVs creates concerns about battery safety that is closely related to its degradation.2–4 Battery degradation reduces the driving range of an EV and could potentially cause catastrophic failures such as thermal runaway.3 While developing new battery electrochemistry, reducing battery degradations through design variable optimization attracts much attention as well.2,6,7

Newman and other researchers have been doing significant work of developing electrochemical battery models for Li-ion batteries (LIBs).8–19 In the meanwhile, the effort has been made on optimizing LIBs design variables using the developed models.12,17,20–25 Electrode thickness, particle sizes, and porosity are considered as key design variables for LIBs.24 Optimizing LIBs design variables has been reported.20–25,29–31 In above studies, the porosity and thickness of the positive electrode are optimized to increase discharge time,25 power and energy density.12,17,22 Both thickness and porosity of electrodes are optimized to maximize specific energy.21 Moreover, both porosity and particle sizes of electrodes are optimized to increase the battery capacity.23 However, the long-term effect of battery degradation, which is affected by the changing of design variables, haven’t been considered in previous studies. This long-term effect can be simulated by applying electrochemical battery models.18,19,28,33–39 Also, few of above studies apply multi-objective optimization algorithms as well as optimizing all the design variables at the same time. Furthermore, an optimization decision is not easy to make as any change of design variable affects battery performance not only in a short period but also over a long term. For example, a thicker electrode can cause more potential drop inside the electrode due to increasing internal resistance. On the other hand, a thicker electrode provides larger volume to hold more active materials (e.g., higher discharge specific power).4,19 Particle sizes of active materials determine the active surface area of electrochemical reactions and the transport of lithium ion in the solid phase. Smaller particles lead to the larger active surface area, which can enable higher reaction flux density (e.g., higher discharge specific power). However, smaller particles may accelerate side reactions such as solid electrolyte interphase (SEI) layer formation26–28 and Manganese (Mn) deposition, which leads to faster degradation. On the other hand, the diffusion of lithium ion is faster in a smaller particle which allows the higher output of current in a short time (e.g., higher discharge specific power).31 Besides electrode thickness and particle sizes, the porosity of the electrode plays an important role in ion transport in the liquid phase. A more porous electrode provides more liquid phase for ion transport. However, the more porous electrode contains less active material, which can affect discharge specific energy. A less porous electrode provides a less liquid phase for ion transport which leads to less discharge specific power and discharge specific energy in the cathode. Therefore, a systematic optimization of the design variables should be used for the complex design problems like battery design. However, few of above studies apply multi-objective optimization algorithms as well as optimizing all the design variables at the same time. Also, the long-term effect of battery degradation, which is affected by the changing of design variables, haven’t been considered in previous studies. Moreover, it is challenging to efficiently solve the optimization problem with multiple design variables and optimization objectives. Furthermore, the computational cost and robustness of the optimization process should be both considered.

In this paper, we develop a multi-objective optimization framework to improve battery performance by optimizing its design variables. The optimization utilizes our previously developed comprehensive capacity fade model that considers most degradations such as SEI layer growth, gas generation, electrolyte oxidation/decomposition, and manganese dissolution/deposition in LIBs. Also, we develop a multi-objective optimization framework considering battery performance and degradation, especially in the long term. Moreover, we develop the guidance for battery design variable optimization. The optimization design aims to find the optimal combination of conductivity, particle sizes, electrode thickness and porosity of both electrodes that can minimize degradation, maximize specific energy, and specific power. A modified Elitist Non-Dominated Sorting Genetic Algorithm (NSGA-II) is utilized for solving the multi-objective optimization that contains seven variables and three optimization objectives via parallel computing.

Methods

Battery degradation model.—The details of the model can be found in our previous paper.40 Tables I and II summarize all governing equations. We constructed all equations in Matlab. The equations were...
discretized and solved via the finite different method in Matlab. The computational domain of the model includes the cathode, separator, and anode, which was divided into uniform grids with a step size of $\Delta x$. The simulation time domain was divided into uniform grids with the step size of $\Delta t$. The implicit central difference method was applied for solving the PDEs. 1D geometry was applied to model the cell level dynamics, which consists of both electrodes and the separator. The 2D geometry was used to model the particle level dynamics, which consists of the radius direction of the spherical particles in both anode and cathode electrodes. The concentration of Li-ion in the

### Table I. A summary of equations of battery model.

| Governor equation | Boundary conditions |
|-------------------|---------------------|
| Mass balance | $\frac{\partial x}{\partial t} = \frac{D}{\Delta x^2} \left( \frac{\Delta x}{\Delta x} \right) + \frac{1}{2} \left( j_{\text{oxid}} + j_{\text{salt}}^1 - 4a_0^1 i_{\text{Mn,react}} \right)$ |
| Charge balance | $\frac{\partial (\varepsilon c_{\text{Mn}})}{\partial t} = \frac{\partial}{\partial x} \left( D \frac{\partial c_{\text{Mn}}}{\partial x} \right) + j_{\text{Mn,react}} + j_{\text{Mn,dep}} = 0$ |
| Electrochemical kinetics | $j_{\text{Mn,react}} = a_0^1 i_{\text{Mn}} \exp[\frac{E_\text{loc, Mn}}{RT} (\Phi_1 - \Phi_2 - R S E I \Phi_2)] - \exp[-\frac{E_\text{F}}{RT} (\Phi_1 - \Phi_2 - R S E I \Phi_2)]$ |
| SOC calculation | $C_{\text{el}} = c_{\text{el}}^l (c_{\text{el}, \text{max}} - c_{\text{el}})^{\alpha_{\text{el}}}$ |

### Table II. A summary of equations of side reactions.

| Side reactions in anode | $i_{\text{Mn,dep}} = -e^{-\delta \alpha_{\text{Mn}} F} \delta_{\text{Mn}} \exp[-\frac{E_{\text{F, Mn}}}{RT} (\Phi_1 - \Phi_2)]$ |
| Side reactions in cathode | $i_{\text{Mn,react}} = -\exp(-\lambda_{\text{SEI}} b) F k_{\text{SEI}} e_{\text{EC}} \exp[-\frac{E_{\text{F, SEI}}}{RT} (\Phi_1 - \Phi_2)]$ |
| Solvent oxidation | $j_{\text{oxid}} = a_0^1 i_{\text{H}} \exp[\frac{E_{\text{F, H}}}{RT} (\Phi_1 - \Phi_2 - U_{\text{oxid}})]$ |
| LiPF$_6$ decomposition | $j_{\text{oxid}} = f_{\text{H}} + j_{\text{oxid}}$ |

$\lambda_{\text{SEI}}$: Reaction rate constant of SEI formation
$F$: Faraday constant
$E_{\text{F, Mn}}$: Mn oxidation potential
$E_{\text{F, SEI}}$: SEI formation potential
$E_{\text{F, H}}$: H oxidation potential
$U_{\text{oxid}}$: Oxidation overpotential
$c_{\text{el}}$: Li-ion concentration
$c_{\text{el}}^l$: Initial Li-ion concentration
$c_{\text{el}, \text{max}}$: Maximum concentration of Li per unit electrode volume
$\delta_{\text{Mn}}$: Mn dissolution rate
$\delta_{\text{H}}$: H dissolution rate
$\lambda_{\text{SEI}}$: Reaction rate constant of SEI formation
$\lambda_{\text{H}}$: H oxidation rate constant
$\lambda_{\text{Mn}}$: Mn dissolution rate
electrolyte, solid phase potential, liquid phase potential, solid phase current density, liquid phase current density, and the pore wall flux were solved in the 1D geometry. The liquid phase potential was set to 0 V as the electric ground at the cathode end. An iteration was required to find the initial solid phase potential to ensure the charge balance (i.e., the total current density of liquid phase and solid phase equals to the applied charge/discharge current density). The pore wall flux from the 1D geometry was projected as the boundary condition of the 2D geometry to solve the Li-ion concentration distribution in the solid particles.

Table I shows equations of Li-ion transport, potential variation in electrodes is treated as mass diffusion in solid spheres. The active particle is assumed to be spherical, and the diffusion of Li-ion in electrodes is treated as mass diffusion in solid spheres. The potential variation in both solid phase φ1 and liquid phase φ2 follows Ohm’s law. The insertion/disinsertion rate of Li-ion is described by Butler-Volmer kinetics.

The schematic diagram of our comprehensive battery degradation model is shown in Figure 1. Table II lists all equations describing LIBs degradations. Side reactions causing battery degradation occur in both electrodes. The simulation parameters for the battery model are listed in Table III and Table IV. In anode, SEI growth, manganese...
deposition, and reduction of H\(^+\) (H\(^+\) is generated from solvent oxidation and salt decomposition) are the main side reactions. As SEI grows thicker, the side reaction rate is slowing down because the electrolyte needs diffuse through the thicker SEI to reach the graphite surface. A decay function \(\exp(-\lambda d)\) is applied to approximate the decaying growth rate with respect to the thickness of the SEI.\(^{40}\) In cathode, dissolution of manganese, solvent oxidation, and salt decomposition are the main side reactions. The side reaction product protons H\(^+\) coming from electrolyte oxidation and LiPF\(_6\) decomposition on the cathode surface, which cause manganese dissolution. Mn\(^{2+}\) and H\(_2\)O are generated as a result of these reactions. The dissolution of manganese plays a key role in cathode capacity fading because of the active material loss, resistance increase, particle structure distortion and isolation.\(^{40}\)

### Elitist Non-Dominated Sorting Genetic Algorithm (NSGA-II).

The NSGA-II is chosen as the optimization algorithm in this study. Genetic algorithm (GA) that simulates the natural evolutionary principle is adopted to identify optimal battery design variables, which requires less information from the optimization problem and is different from the classical optimization algorithms such as direct method and gradient-based method. Elite-preserving operators are introduced in the NSGA-II for minimizing cost functions.\(^{41}\) The NSGA-II algorithm passes the elites (i.e., the best individuals in each generation) of a population down to the new generation. Therefore, it preserves good results in the early runs until a better solution is found. The population described herein means individuals with different combinations of decision variables. Battery design variables are decision variable \(x\). We choose the porosity, thickness and particle size in both electrodes as six design variables. Also, the electronic conductivity in the cathode is considered as the seventh design variable. The reason that the electronic conductivity in the anode is not taken into account is because of its high conductivity. Thus, seven design variables are considered in this study. The constraints of each design variable \(x\) are listed in Table V. The generation represents all the population in one iteration.

### Table IV. Side reaction parameters.

| Parameter | Value               | Parameter | Value               |
|-----------|---------------------|-----------|---------------------|
| \(k_{SEI}\) | \(1 \times 10^{-9} \text{ m} \cdot \text{s}^{-1}\) | \(a_{\text{Mn}^{2+}}\) | 0.5                 |
| \(c_{EC}\)  | 4541 \(\text{mol} \cdot \text{m}^{-3}\) | \(U_{\text{red}}\) | 4.1 V               |
| \(k_{SEI}\) | 4 \times 10^{-2} \text{ m} \cdot \text{s}^{-1}\) | \(f_{\text{H}^{+}}\) | 15%                |
| \(a_{\text{Mn}^{2+}}\) | 0.162 \(\text{kg} \cdot \text{mol}^{-1}\) | \(k_{\text{dec}}\) | 40 \(\text{A} \cdot \text{m}^{-3}\) |
| \(M_{SEI}\)  | 1690 \(\text{kg} \cdot \text{m}^{-3}\) | \(D_{\text{f}^{+}}\) | \(7.13 \times 10^{-10} \text{ mol} \cdot \text{m}^{-2} \cdot \text{s}^{-1}\) |
| \(\rho_{SEI}\) | 1 \times 10^{-5} \text{ m} \cdot \text{s}^{-1}\) | \(D_{\text{f}^{+}}\) | \(1 \times 10^{-11} \text{ m}^{2} \cdot \text{s}^{-1}\) |
| \(k_{\text{Mn},\text{dep}}\) | 0.5 | \(D_{\text{f}^{+}}\) | \(5 \times 10^{-11} \text{ m}^{2} \cdot \text{s}^{-1}\) |
| \(a_{\text{Mn},\text{dep}}\) | 5 \times 10^{5} \text{ m}^{-1}\) | \(V_{\text{f}^{+}}\) | 3 \times 10^{-11} \text{ m}^{2} \cdot \text{s}^{-1}\) |
| \(k_{\text{H}^{+}}\) | 1 \times 10^{8} \text{ m}^{-1}\) | \(k_{\text{iso}}\) | 220                 |
| \(a_{\text{H}^{+}}\) | 2.07 \times 10^{-6} \text{ m} \cdot \text{s}^{-1}\) | \(k_{\text{cso}}\) | 1.4 \times 10^{-4} \text{ m} \cdot \text{mol}^{-1}\) |
| \(k_{\text{di}}\) | 0.5 | \(k_{\text{cso}}\) | 3 \times 10^{4} \text{ \Omega} \cdot \text{m}\) |
| \(k_{\text{di}}\) | 5 \times 10^{-3} \text{ m} \cdot \text{s}^{-1}\) | \(T_{\text{di}}\) | 80%                |

aCalibrated values.  
bFrom reference 40.

The four steps of optimization are summarized in the flowchart, as shown in Figure 2. In 1\(^{st}\) step, NSGA-II starts by randomly generating the parent population \(P_t\) with size \(N\) among all the possible decision variables \(x\). Then, the offspring population \(Q_t\) is generated from the \(P_t\) with the same size \(N\). The 2\(^{nd}\) Step is followed by combining \(P_t\) and \(Q_t\) together to form a population \(R_t\) with size \(2N\). \(R_t\) is evaluated by a non-dominated sorting, which is an algorithm that sorts a population into an ascending level of non-domination. Then in the 3\(^{rd}\) step, the

### Table V. Design variables and the ranges.

| Design variables     | Lower bound | Upper bound |
|----------------------|-------------|-------------|
| Cathode particle radius (um) | 1           | 20          |
| Anode particle radius (um)   | 1           | 20          |
| Cathode porosity   | 0.1         | 0.4         |
| Anode porosity     | 0.1         | 0.4         |
| Cathode thickness (um)   | 50          | 150         |
| Anode thickness (um)   | 50          | 150         |
| Cathode conductivity (S m\(^{-1}\)) | 1          | 10          |

Figure 2. Flowchart of NSGA-II optimization.
solution of the non-dominated sorting forms the new parent population \( P_{t+1} \) with the size of \( N \). In the 4th Step, an offspring population \( Q_{t+1} \) is created from \( P_{t+1} \) by using the elite-preserving operators like, crowded tournament selection, crossover and mutation to keep the diversity. 2nd to 4th Steps are repeated until the stopping criterions are satisfied. Maximum generations of 300 and function tolerance of 0.001 are set as the stopping criterions. When the stop criterions are satisfied, the optimization stops and gives Pareto-optimal solutions. The purpose of this optimization study is to improve LIBs for EVs application, which requires instant power for acceleration, sufficient travel range per charge, and minimized capacity loss over time. We set maximizing discharge specific power, discharge specific energy, and battery capacity as three optimization objectives, and their objective functions are defined as follows,

Maximize discharge specific energy
\[
\text{Maximize discharge specific energy} \quad \text{Max} \left\{ E_{\text{cell}} = \frac{1}{M_{\text{cell}}} \int_{0}^{\text{discharge-time}} V_{\text{cell}}(t) I dt \right\} \quad [1]
\]

Maximize discharge specific power
\[
\text{Maximize discharge specific power} \quad \text{Max} \left\{ P_{\text{cell}} = \frac{1}{10 \cdot M_{\text{cell}}} \int_{0}^{10} V_{\text{cell}}(t) I dt \right\} \quad [2]
\]

Maximize capacity at the end of cycles
\[
\text{Maximize capacity at the end of cycles} \quad \text{Max} \left\{ Q_{\text{cell}} = \frac{1}{M_{\text{cell}}} \int 1 dt \right\} \quad [3]
\]

Design variable \( x \) subject to \( x_{\text{lower}} < x < x_{\text{upper}} \)

Cell mass \( M_{\text{cell}} = M_{\text{case}} + M_{c} + M_{a} + M_{\text{sep}} \quad [4] \)

**Results and Discussion**

**Model validation.**—We developed a three-electrode cell for model validation, as shown in Figure 3. The cell is made of graphite as the anode, LiMn_2O_4 as the cathode, and lithium metal as the reference electrode. In order to avoid the electroplating effect, \(^{42,43}\) the anode is placed in the coin cell positive case, and the cathode is placed into the coin cell negative case. The reference electrode adopted herein is used for measuring potentials in anode and cathode, respectively. The reference electrode is connected with a wire that extends to outside the battery. The wire is insulated and protected by Kapton tape to avoid short circuit by touching the positive or negative cases. The assembled cell is tested by using an Arbin battery cycler for 270 cycles and 470 cycles, respectively. The charge and discharge rate is set as 1C. The constant current constant voltage (CCCV) method is applied to charge and discharge the battery. When the cutoff voltage 3.4 V/4 V is reached, the voltage is held until current decreases to 0.1C. The electrode potentials are recorded during cycling. The battery model is simulated under the same condition as the experiment. The parameters that are used for modeling are listed in Tables III and IV. For the 270-cycle test, the experiment took more than 540 hours while the computational cost of the simulation is around 1.5 hours. In Figure 4, the experiment and simulation results are compared. The potentials over long term cycling in Figures 4a and 4b show two different stages. In the first stage, the potentials from both electrodes have a slightly increasing trend. In the second stage, the potentials decrease gradually. Moreover, the band of cathode potential becomes wider over cycles, and the band of anode potential becomes smaller over cycles. These changes are caused by side reactions such as the fast growth of SEI...
layer at the beginning of the battery lifetime and the slow growth of SEI layer at the second stage when other degradations become more significant. Overall, it can be seen that the lower bound of the cathode potential extends from 3.8 V to 3.6 V and the upper bound of the anode potential drops from 0.35 V to 0.26 V. The battery model is calibrated with experimental measurements. Then, the calibrated battery model is utilized for battery design variable optimization.

Here we propose a water-glass analogy to explain the shift of SOC as shown in Figure 5. If we consider SOC of a battery is analogous to the level of water in a glass cup. SOC equals 1 equivalent to a full cup of water and SOC equals 0 equivalent to an empty cup. The level of water indicates the remaining water in the cup. Similarly, we use the value of SOC between 0 and 1 to indicate the remaining energy in the battery. In reality, the size of the cup doesn’t change over time. Hence if the same amount of water is removed and added from the cup, the level of water will be the same as before. However, the battery capacity decreases over time due to the degradations. In this case, the size of the cup shrinks over time. Therefore, the level of water will increase if the same amount water is discharged and charged to the cup. This explains the increasing upper bound of SOC in both electrodes over time. Also, the side reactions contribute to the shift of SOC. The lithium flux $j_{Li}$ of mass balance in Table I contains side reaction terms. The various value of side reaction terms can increase or decrease $j_{Li}$, which significantly affects the surface concentration of $c_x$. As a result of surface concentration change, the rate of SOC is affected. Overall, the SOC shift is caused by the combination of battery capacity loss and side reactions due to battery degradation.

The optimization of battery design variables.— Six Pareto-optimal solutions (i.e., set 1 to 6) are presented in Figure 6, where each corner represents a design objective. The values of each optimization objective are normalized from 0 to 1 and are marked as the dark gray triangle lines between the corners and the center of the chart.

Figure 5. Water-cup analogy. In (a), cathode capacity decrease causes the SOC swinging. The variation of SOC ranges leads to the change of cathode potential. In (b), the anode potential adjusts itself to match the cutoff voltage.
Table VI. Pareto-optimal solutions.

| Set | Porosity | Radius [μm] | Conductivity [Ω m] | Thickness [μm] | Porosity | Radius [μm] | Thickness [μm] |
|-----|----------|-------------|--------------------|----------------|----------|-------------|----------------|
| 1   | 0.10     | 1.2         | 10                 | 75             | 0.11     | 13.4        | 50             |
| 2   | 0.10     | 2.3         | 10                 | 75             | 0.10     | 12.7        | 50             |
| 3   | 0.19     | 10.2        | 9                  | 89             | 0.29     | 14.8        | 100            |
| 4   | 0.23     | 1           | 10                 | 102            | 0.30     | 15.0        | 109            |
| 5   | 0.20     | 18.4        | 6                  | 109            | 0.39     | 15.5        | 134            |
| 6   | 0.24     | 13.4        | 3                  | 144            | 0.39     | 15.5        | 134            |

Figure 6. The objective values of each design set are presented in solid colored lines. The corresponding optimized battery design variables of each solution are listed in Table VI. It shows that set 2 has the lowest capacity of 0.19 and set 6 has the highest capacity of 0.9. If we compare the design variables of set 2 and set 6 in Table VI, set 6 has larger porosity and thickness as well as larger particle size in both electrodes. In term of discharge specific energy, set 2 has the highest discharge specific energy of 0.9 and set 4 has the lowest discharge specific energy of 0.55. As seen from Table VI, set 2 has the smallest porosity and thickness among all the solutions. Therefore, we can draw the following conclusion that porosity and thickness are closely related to capacity and discharge specific energy. Larger porosity, thickness and particle size lead to a higher capacity. Smaller porosity and thickness increase discharge specific energy. If we compare Equations 1 and 3, cell output voltage U is considered in discharge specific energy calculation. Smaller porosity and thickness lead to higher cell voltage so that discharge specific energy is higher. In Figure 5, the discharge specific power shows its highest value in set 4 and the lowest value in set 5 and set 3. It can be seen from set 4 in Table VI that the discharge specific power is related to electrode thickness, particle sizes, conductivity, and porosity. A thinner anode electrode can shorten the path of lithium ion transport; a thicker cathode electrode ensures enough space for receiving Li-ion; a larger porosity in cathode electrode provides more liquid phase for ion transport; smaller particles increase the surface area and reduce the path of solid phase diffusion.

Considering the highest discharge specific power within six sets of Pareto-optimal solutions, we pick the battery design set 4 for 750 cycle’s simulation to compare the performance of the battery before and after optimized design, as shown in Figure 7. Figures 7a and 7c are the results before the optimization. Figures 7b and 7d are the results after the optimization. The anode SOC is between 0.01–0.55 in Figure 7a. The cathode SOC has a range of 0.55–0.99 in Figure 7c. The SOCs in both Figures 7a and 7c have a slight drop at the beginning and gradually shift upward over time. The shift of SOC windows increases the complexity of battery control strategy development. Comparing with the results before the optimization, the SOCs in Figures 7b and 7d do not have a significant upward shift like the ones in Figures 7a and 7c.

Figure 8 shows the comparison of SEI layer growth before and after optimization. The Pareto-optimal solution set 4 (see Table VI) was adopted herein to reduce electrodes thicknesses, particle sizes, and porosity. The simulations results show that 50% reduction of the SEI thickness after optimization (i.e., reducing from 200 nm to 105 nm after 750 cycles). Both SEI layers rapidly grow during the first 50 cycles. However, the optimized cell has a smaller SEI growth rate. By comparing the design parameters between before and after optimization in Table VI, the optimized cell has the significant smaller porosity and thickness in the anode. Both porosity and thickness affect the current and potential distribution during charge/discharge and are one of the reasons optimized design has less SEI growth during cycling.

Figure 9 shows the spatial dependent SEI thickness of the anode in Figure 9a and residual cathode capacity in Figure 9b. The electrode length in both plots is normalized from 0 to 1. In Figure 9a the SEI thickness is plotted vs. the anode length. We can observe from this
Figure 8. 3D plot of SEI layer growth over 750 cycles. Here, the “0” along anode length means near separator and the “1” means near current collector. The SEI layer reduces 50% after optimization (i.e., the Pareto-optimal solution set 4, see Table VI).

The results in Figure 11 indicates that each design variable has its unique influence on battery’s discharge specific energy, discharge specific power, and capacity. In each sub figure, there is one design variable that changes in ascending order, which is plotted as the black dots in the 3D space. In the meanwhile, other design variables keep as constants. If a large discharge specific energy is required, the electrodes need to be thin in anode and thick in cathode, referring to Figures 11a and 11b; particles need to be smaller in cathode electrode, referring to Figures 11c and 11d; the electrode need to be more porous in cathode and less porous in anode, seen in Figures 11e and 11f. This is because a thin anode electrode can shorten the path of lithium ion transport, a thicker cathode electrode can store more Li-ion; smaller particles increase the surface area as well as reducing the path of solid phase diffusion; a larger porosity in cathode electrode provides more liquid phase for ion transport. If a larger capacity is required, the electrodes need to be thick in both
Figure 11. 3D plots of sensitivity analysis of design variables. Three axes represent three objectives. Each design variable changes from small to large, the corresponding results are plotted in the 3D space with black dots. The trajectory of the black dots is projected to three planners. The projection of the red dots shows the plot of specific power vs. capacity; the projection of the blue dots shows the plots of specific energy vs. capacity; the projection of the pink dots shows the plots of specific energy vs. specific power.

Conclusions

A battery design optimization framework is developed in this paper to improve battery performance and reduce degradation over time. The multi-objective optimization problem has three objectives and seven design variables. The degradation battery model is served to output objective values: discharge specific power, discharge specific energy and residual capacity with the inputs of design variables: thickness, porosity, particle radius, and conductivity. Several conclusions can be drawn from this paper:

1. The validated degradation battery model can simulate battery long term performance with considering spatial dependent degradations in both electrodes.
2. The complex design optimization problem with seven design variables and three objectives is successfully solved by the NSGA-II. By applying parallel computing with this algorithm, the computational cost of the optimization process is significantly reduced.
3. The obtained Pareto-optimal solutions are obtained from NSGA-II. From the comparison, the optimized cell has better performance over 750 cycle’s simulation: less SOC swing and less reduction of discharge specific energy. The improvement is favored by the battery management system of EVs.
4. The battery degradation of SEI growth and residual cathode capacity are also studied after 750 cycles. The average SEI growth of the optimized cell is 95 nm less than the before optimization. Moreover, the optimized cell has 6.5% higher average residual capacity in the cathode. Furthermore, both SEI...
growth and residual capacity are much more uniform in the opti-
mized cell, which is considered a more effective usage of the
battery.
5. The water-cup analogy is first time proposed by this work to
explain the battery degradation induced SOC shifting during long-
term cycling test.
6. The design optimization framework can also be used for various
type of batteries and applications. Different objectives and design
variables can be used for specific applications.

List of Symbols

- $\alpha_a^H$: Active surface area per anode electrode unit volume
- $\alpha_c^H$: Active surface area per cathode electrode unit volume
- $A_D$: Electrode area ($cm^2$)
- $C_D$: Double-layer capacitance ($F m^{-2}$)
- $c$: Concentration in solid phase ($mol m^{-3}$)
- $c_\ell$: Concentration in liquid phase ($mol m^{-3}$)
- $c_{EC}$: Solvent concentration ($mol m^{-3}$)
- $C$: Battery capacity ($Ah$)
- $C_n$: Current density of solid phase ($A m^{-2}$)
- $C_m o l$: Molmolar
- $C_{mol}$: Molar concentration
- $D_{cell}$: Initial battery capacity ($Ah$)
- $C_1$: Initial battery capacity ($Ah$)
- $C_2$: Coefficient
- $E_{cell}$: Specific energy ($kW kg^{-1}$)
- $D_{EC}$: Diffusion coefficient of solvent ($m^2 s^{-1}$)
- $D_2$: Diffusion coefficient of lithium species in liquid phase
- $D_s$: Diffusion coefficient of lithium species in solid phase
- $D_{eff}$: Effective electrolyte phase Li diffusion coefficient
- $D_{eff}$: Effective solid phase Li diffusion coefficient
- $D_{eff}$: Effective diffusion coefficient of $H^+$
- $D_{eff}$: Effective diffusion coefficient of $Mn^{2+}$
- $D_{eff}$: Effective diffusion coefficient of $H_2O$ ($m^2 s^{-1}$)
- $F$: Faraday’s constant, 96487 (C $mol^{-1}$)
- $f_{H^+}$: Fraction coefficient for $H^+$ dissolution
- $f_{Mn\...}$: Fraction coefficient for $Mn^{2+}$ dissolution
- $f_h$: Electrolyte activity coefficient
- $I$: Applied current ($A$)
- $i_{pot}$: Exchange current density of the solvent oxidation reaction
- $i_{Mn\...}$: Current density of Mn reaction
- $i_{SEI}$: Current density of SEI reaction
- $i_{Mn\...}$: Current density of Mn deposition
- $i_{H2}$: Current density of $H_2$ generation
- $i_{ox}$: Current density of solid phase oxidation
- $i_{oc}$: Local reaction current density
- $i_{red}$: Reaction current density of side reaction ($A m^{-3}$)
- $i_{Li}$: Reaction current density of total Li intercalation or de-
tercalation ($A m^{-3}$)
- $k_{SEI}$: Reaction coefficient of salt decomposition
- $k_{acid}$: Reaction coefficient of acid attack on the active material
- $k_{H2}$: Reaction coefficient of $H_2$ generation
- $k_{iso}$: Reaction coefficient of particle isolation coefficient
- $k_{Mn\...}$: Reaction coefficient Mn deposition
- $k_{SEI}$: Reaction coefficient of SEI ($m s^{-1}$)
- $k_{H2}$: Reaction rate coefficient of intercalation
- $k_{Li}$: Reaction rate coefficient of side reaction ($m s^{-1}$)
- $L$: Cell width ($m$)
- $M_{cell}$: Cell mass ($kg$)
- $M_{sep}$: Mass of separator ($kg$)
- $M_{case}$: Mass of case ($kg$)
- $M_a$: Mass of anode ($kg$)
- $M_c$: Mass of cathode ($kg$)
- $M_{SEI}$: SEI molar mass ($kg mol^{-1}$)
- $P$: Specific power ($W kg^{-1}$)
- $P_{req}$: Requirement of specific power ($W kg^{-1}$)
- $Q_{cell}$: Cell capacity ($Ah$)
- $R$: Gas constant ($Jmol^{-1}K^{-1}$)
- $R_a$: Radius of active particle (A)
- $R_{SEI}$: SEI resistance ($\Omega m^2$)
- $t$: Time ($s$)
- $\Delta t$: Time step
- $U_a$: Anode electrode equilibrium potential
- $U_c$: Cathode electrode equilibrium potential
- $U^{red}_{oxid}$: equilibrium potential of solvent oxidation
- $\tau$: Time ($s$)
- $\Delta \tau$: Time step
- $\chi$: Battery design variables
- $\bar{V}$: Molar volume of LMO ($m^3 mol^{-1}$)

Greek

- $\rho_{SEI}$: Cathodic charge transfer coefficient for $H_2$ generation
- $\alpha_{Mn\...}$: Cathodic charge transfer coefficient for Mn deposition
- $\alpha_{SEI}$: Cathodic charge transfer coefficient for SEI formation
- $\alpha_{Mn\...}$: Anodic charge transfer coefficient for solvent oxidation
- $\alpha_{Li}$: Anodic charge transfer coefficient for lithium intercalation or deintercalation
- $\alpha$: Cathodic charge transfer coefficient for lithium intercalation or deintercalation
- $\sigma_{eff}$: Effective solid phase conductivity ($S m^{-1}$)
- $\sigma_{eff}$: Effective liquid phase conductivity ($S m^{-1}$)
- $\beta$: Solid phase potential ($V$)
- $\gamma$: Liquid phase potential ($V$)
- $\eta$: Overpotential ($V$)
- $\delta$: SEI layer thickness ($nm$)
- $\epsilon$: Cathode active material volume fraction
- $\epsilon$: Cathode electrolyte phase volume fraction
- $\epsilon$: Anode active material volume fraction
- $\epsilon$: Anode electrolyte phase volume fraction
- $\lambda_{Mn\...}$: Limiting coefficient of Mn deposition ($m^{-1}$)
- $\lambda_{SEI}$: Limiting coefficient of SEI reaction ($m^{-1}$)
- $\lambda_{H2}$: Limiting coefficient of $H_2$ generation ($m^{-1}$)
- $\beta$: SEI layer porosity
- $\rho_{SEI}$: SEI density ($kg m^{-3}$)

Subscripts

- $s$: Solid phase
- $l$: Liquid phase

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