Heuristic Versus Optimal Charging of Supercapacitors, Lithium-Ion, and Lead-Acid Batteries: An Efficiency Point of View

Yasha Parvini, Ardalan Vahidi, and S. Alireza Fayazi

Abstract—Electrical energy storage systems are extensively utilized in applications, including electrified vehicles, renewable power generation, and electronic devices. While discharging events are a function of the power demand, the charging procedure is often controllable. This paper evaluates different charging strategies for stand-alone supercapacitors (SCs), lithium-ion (Li-ion), and lead-acid batteries. Constant power and optimal charging strategies are formulated and the corresponding charging currents are obtained. Efficiency analysis for different charging strategies and charging times (slow and fast) is performed. The identical objective function for all modules is to minimize the resistive losses during a given charging time by utilizing Pontryagin’s minimum principle. An analytical solution exists for the SC case, which is constant current charging. The variation of the total internal resistance with state of charge in lead-acid chemistry is considerable, and the optimal charging problem results in a two-point boundary value problem. In case of the Li-ion battery, the model includes the electronic as well as polarization resistance. Furthermore, in order to investigate the influence of temperature on the optimal charging of the Li-ion battery, a constrained optimal control problem for a three state electrothermal model is formulated and solved using dynamic programming.

Index Terms—Battery, efficiency, energy storage, fast charging, optimal control, supercapacitor (SC).

I. INTRODUCTION

Batteries have become an indispensable part of our daily life. They can be found almost everywhere from powering our electronic gadgets, computers, and phones to electrifying our vehicles and also form a critical part of the modern centralized and distributed power grids. Supercapacitors (SCs), on the other hand, are the premier energy storage devices in terms of power density, long cycle life, and the ability to operate at extreme temperatures. Much research and development are spurred toward studying important factors influencing the efficiency and cycle life of batteries and SCs, such as monitoring and control of the cell charging method, current rate, number of charging/discharging cycles, and temperature [1]–[10].

Studies that investigate the optimization of efficiency over the entire driving cycle mask the fundamental bottlenecks of efficiency in the electrical energy storage systems. In stand-alone operation and during discharge, the cycle is often imposed by the required load, and therefore, there is little that can be done in reducing resistive losses. During charging, however, there is the opportunity to choose the charging time and profile, such that resistive losses are reduced. Battery manufacturers often have a recommended charging profile, which may be suboptimal.

There are numerous studies focusing on different charging methods to achieve objectives, such as decreasing the charging time [11]–[15], life span [16]–[20], and efficiency maximization or cost minimization. Optimal charging of lithium-ion (Li-ion) batteries is studied in [21], where minimizing the charging time while satisfying specific physical and thermal constraints is considered. In [22], an optimal charging problem is solved for an SC during regenerative braking with the objective of minimizing ohmic losses. Suthar et al. [23] use a single-particle model and aim to find the optimal current profile, with the objective of maximizing the charge stored in the cell in a given time and with the constraint of minimal damage to the electrode particles during intercalation. Bashash et al. [24] focus on optimizing the timing and charging rate of a plug-in hybrid electric vehicle from the power grid where the goal is to simultaneously minimize the total cost of fuel and electricity and the total battery health degradation. Optimizing the battery charging power in photovoltaic battery systems is studied in [25], where different objectives, such as charging time, battery life time, and cost of charging, are considered. Inoa and Wang [26] suggest optimal charging profiles in order to minimize charging losses and reach a preset temperature at the end of the charging time for Li-ion batteries. More recently, [27] and [28] have solved the optimal charging problem for Li-ion batteries considering the tradeoff between charging time and energy loss in the objective function; however, neither of these papers consider the transient effect of temperature on electrical model parameters of the Li-ion battery. One important output of the mentioned studies is the charging/discharging current profile that satisfies the specific objective functions. For example, there are studies with the objective of reducing the charging time, which
result in different types of constant current (CC) charging methods, such as multistage charging [29]–[33], impedance compensation [34]–[36], and pulse charging [37]. Constant voltage (CV) charging has been used to improve charging speed by combining the battery pack and charger models and the results are compared with the CC method [38]. Constant power (CP) charging and discharging are also of interest to researchers as it corresponds to real-world operating conditions, such as in hybrid and electric vehicles. Modeling the thermal behavior of Li-ion batteries under CP charging and discharging cycles is investigated in [39]. Under CP, the relationship between the available energy in a battery and charging power has been investigated in [40]. The aging of SCs under CP charging condition is studied in [41].

The distinctive aspects of this paper compared with the preceding literature are as follows.

1) Our approach isolates the storage system and studies its charging as a component. Unlike system level studies, the charging bottlenecks of the module in this method are clearly identifiable. The significance of this approach is that the results, regardless of the application, are universal.

2) Unlike generic models used in the literature, the model of each module is developed through experimental parameterization in separate studies by the authors and their colleagues. This increases the reliability of the charging profiles and the efficiency analysis.

3) This paper aims to use simple models to break down the problem into cases where the effect of every model parameter on the final result can be identified clearly. This approach also allows to use analytical methods in a number of scenarios. The nature of this paper sets the stage for future studies that attempt to use more complex models.

4) The comparison of almost all charging strategies that are widely used in practice (CV, CC, CP, and CCCV) with their optimal charging scenario counterpart introduces a justifiable framework for choosing the right strategy in a given application. Furthermore, the effect of reduced charging time (fast charging) on the results of the heuristic as well as optimal strategies is investigated.

5) As the most widely used storage technology in transportation and electronic devices, the Li-ion battery is chosen for this paper with a gradual increase in model complexity throughout this paper. The effect of shorter charging times and the resulting increase in temperature, a potential cause of thermal runaway, is investigated in this paper. Discussion on SC charging is included in this paper, which unlike the rich battery literature, is not widely studied.

The common considerations in the problem formulation and analysis for the three SCs, lead-acid, and Li-ion modules are as follows.

1) In all optimal charging formulations, the objective function is to maximize the charging efficiency by minimizing the resistive losses in a given charging time and for a specified range of state of charge (SOC).

2) All problems are formulated using Pontryagin’s minimum principle (PMP) method, with the intention to solve them analytically. In cases where analytical solution is not feasible, the problem is solved using numerical methods.

3) The CP and optimal charging current profiles and efficiencies are obtained and compared for all modules.

4) Both slow and fast charging times are investigated.

We begin with lumped models for each module with the upper and lower bounds on SOC being the only constraint considered in the optimal control problem formulations. Later on, we solve the optimal charging problem for the Li-ion battery by coupling a reduced order, two state thermal model to the electrical model in order to investigate the effect of temperature on the optimal result. In the electrical models used in this paper, $R_1$ indicates the ionic and electronic resistance of electrolyte and also the electronic resistance of the electrode. The other two parameters are the charge-transfer resistance $R_1$, which is in parallel with the double layer capacitance $C_1$ formed at the interface between the electrode and the electrolyte [42]. In this paper, the sum of $R_1$ and $R_2$ is called the total internal resistance $R$. These validated equivalent electric circuit models guarantee the robustness of the model parameters. Also such models facilitate the application of analytical methods that produce results with the highest reliability. However, in order to consider real-world limitations, such as temperature constraints, more complex models, such as the electrothermal model used in this paper, are more suitable. Furthermore, high fidelity electrochemical–thermal models have the capability of clarifying the microscopic bottlenecks in the charging process, such as the lithium plating at high currents and low temperatures. In applications were the charging is performed off-line (i.e., charging an electric vehicle through the outlet) with longer charging times, using simple models, such as the ones in this paper, will be sufficient. In conditions where the goal is to minimize the charging time, the utilization of more complex models is suggested.

In case of the SC, the electrical dynamics is modeled using a constant total internal resistance $R$. The open circuit voltage (OCV) is assumed to have a linear relationship with SOC. Another assumption is constant ambient temperature of 25 °C during charging. Due to high power density of SCs, the fast and slow charging times are chosen to be 30 s and 6 min, respectively. The optimal charging current and efficiency for the SC have analytical solutions and are compared with the CC and CV strategies. For the lead-acid battery, similar to the SC, the module is modeled using a total constant internal resistance $R$ at a constant ambient temperature of 25 °C. The difference in the problem formulation for the lead-acid battery is the strong dependence of $R$ on SOC, which is integrated into the electrical model. Both OCV and $R$ are approximated by the second-order polynomials as a function of SOC. The fast and slow charging times are chosen to be 6 min and 1 h, respectively. Considering these conditions for the lead-acid battery, the optimal charging formulation results in a two-point boundary value problem (TPBVP), which is solved numerically. The obtained optimal charging current and efficiency are compared with CP and CC methods.

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Li-ion battery, in the first step, only the electronic resistance \( R_s \) with a constant value at a constant charging temperature of 25 °C is considered. In the second step, the effect of charge-transfer resistance \( R_t \) and double layer capacitance \( C_1 \) is added to the model. The dependence of \( R_s \) on \( SOC \) is negligible at room temperature, and the variation of \( R_t \) and \( C_1 \) on \( SOC \) has not been considered. Similar to the lead-acid battery, the charging times studied are 6 min and 1 h for rapid and slow charging strategies, respectively. Finally, in order to investigate the effect of temperature on the electrical characteristics, a validated three state electrothermal model is utilized and the optimal charging problem is solved using the numerical method of dynamic programming (DP). The optimal solution in this case is obtained subject to temperature constraints. The unconstrained optimal charging scenarios were partially reported in [22] and [43].

The remainder of this paper is organized in the succeeding order. Section II focuses on the SC, including the model used, CV, CP, optimal charging strategies, and the efficiency analysis. Section III explains the model, charging, and efficiency analysis of the lead-acid battery. Section IV describes the model, charging, and efficiency analysis of the Li-ion battery. Section V presents the electrothermal model and the optimal charging of the Li-ion battery, considering the temperature effect as well as the voltage and temperature constraints. Section VI presents the conclusion remarks.

II. CHARGING OF THE SUPERCAPACITOR

A. Supercapacitor Model and Specifications

The SC utilized in this paper is a Maxwell BCAP3000 cylindrical double layer cell. The specifications of the cell are listed in Table I.

The nominal energy capacity of the SC according to Table I is 3 Wh. The equivalent electric circuit model for this cell is identified using pulse-relaxation experiments for a wide range of temperatures from −40 °C to 60 °C in [44] and [45]. The model indicates that the dependence of the model parameters, such as \( R \) in Fig. 1 on \( SOC \) and also the current magnitude is negligible. Also as mentioned in Section I, we do not include the thermal constraints in the charging problem formulation and efficiency analysis for the SC. The value of constant electronic resistance \( R \) is 2.97 mΩ for this cell at 25 °C. The OCV profile as a function of \( SOC \) is assumed to be linear.

B. Constant Voltage Charging of the Supercapacitor

It is well known that charging a capacitor and similarly an SC, from zero charge to full charge, with a CV source results in 50% energy loss, irrespective of the internal and line resistances. This can be easily shown by writing the differential equation governing the SC’s stored charge \( q(t) \), for the circuit shown in Fig. 1

\[
R \frac{dq}{dt} + \frac{q}{C} = V_{dc}
\]

where \( C \) is the nominal capacitance, \( R \) is the total internal resistance, and \( V_{dc} \) is the charging voltage. If \( V_{dc} \) remains constant over time, the solution to the above-mentioned differential equation from a zero initial charge condition can be obtained as

\[
q(t) = CV_{dc} \left[ 1 - e^{-t/RC} \right]
\]

and the CV charging current \( I_{cv} \) is then

\[
I_{cv}(t) = \frac{V_{dc}}{R} e^{-t/RC}.
\]

The resistive energy loss is obtained by integrating the resistive power loss \( R I_{cv}^2 \) over the entire charging interval \([0, +\infty)\) as

\[
E_{loss,cv} = \frac{V_{dc}^2}{R} \int_0^{\infty} e^{-2t/RC} dt = \frac{1}{2} CV_{dc}^2.
\]

This amount is equal to the total energy stored in the SC. In other words, the efficiency of charging an empty SC with a CV source is 50%, independent of resistance \( R \). Note that the charging efficiency depends on both the initial and final SOC. For example, charging an SC from half full charge with CV has an efficiency of 75%. Using the definition of efficiency, charging an SC from an initial SOC (\( SOC_i \)) to a final SOC (\( SOC_f \)) with a CV source is

\[
\rho_{cv} = \frac{1}{1 + \frac{(1 - SOC_f)^2}{SOC_i^2 - SOC_f^2}}.
\]

C. Constant Power Charging of the Supercapacitor

Consider charging the SC with the model in Fig. 1 by replacing the CV source with a CP source of \( P_0 \). Applying Kirchhoff’s voltage law to the circuit the stored charge dynamics is

\[
R \frac{dq}{dt} + \frac{q}{C} = \frac{P_0}{q} \rightarrow \dot{q} = I_{cp} = \frac{-q \sqrt{\frac{q}{2}} + \sqrt{\left(\frac{q}{2}\right)^2 + 4RP_0}}{2R}
\]

where \( I_{cp} \) is the CP charging current. Equation (6), which is a nonlinear differential equation, can be solved numerically to find the charge and current. Consider charging the cell from zero to full charge. Figs. 2 and 3 show the charge stored in...
This is an optimal control problem and can be solved using PMP method [46]. First, form the Hamiltonian

$$H(x_1, u_1, t) = Ru^2(t) + \lambda_1(t) \frac{u(t)}{CV_{\text{max}}}$$  \hspace{1cm} (9)$$

where $\lambda_1$ is a costate. The optimal costate should satisfy the subsequent dynamic equation

$$\frac{d}{dt} \lambda_1(t) = -\frac{\partial H}{\partial x_1} = 0$$ \hspace{1cm} (10)$$

implying that, in this specific problem, the optimal $\lambda_1$ must be a constant. The unconstrained optimal solution will also need to satisfy the condition

$$\frac{\partial H}{\partial u} = 0 \rightarrow u(t) = -\frac{1}{2} \frac{1}{RCV_{\text{max}}} \lambda_1(t)$$ \hspace{1cm} (11)$$

showing that the optimal input (charging current) must be a constant. At this point, we can integrate (7) and use the boundary conditions $x_1(0) = SOC_1$ and $x_1(t_f) = SOC_f$ to find the value of this optimal and constant input

$$u_{\text{opt}}(t) = I_{\text{opt},SC} = \frac{CV_{\text{max}}(SOC_f - SOC_1)}{t_f}$$ \hspace{1cm} (12)$$

where the subscript “opt” denotes the optimal solution. This is, in fact, the minimizing solution, since $(\partial^2 H / \partial u^2) > 0$. Given a specific charging time, the most efficient way to charge the SC will be applying a CC equal to (12). The optimal charging current is a CC of 22.5 and 270 A for charging the SC from zero charge to full charge in 6 min and 30 s, respectively.

E. Efficiency Analysis for the Supercapacitor

In order to obtain the charging efficiency of the SC, the total storable energy is required. By the integration of power over the entire charging time and using the definition of SOC, the total energy stored in the SC is obtained as

$$E_{\text{sc}} = \frac{1}{2} CV_{\text{max}}^2 [SOC_f^2 - SOC_1^2].$$ \hspace{1cm} (13)$$

The energy loss in the SC during optimal charging is already known and is equal to $\int_0^{t_f} Ru_{\text{opt}}^2(t) dt$. Then, the optimal charging efficiency is

$$\rho_{\text{opt}} = \frac{1}{2} CV_{\text{max}}^2 (SOC_f^2 - SOC_1^2) - \int_0^{t_f} Ru_{\text{opt}}^2(t) dt.$$

Substituting for $u_{\text{opt}}$ from (12) yields

$$\rho_{\text{opt},SC} = \frac{1}{1 + \left( \frac{2RC}{t_f} \right) (SOC_f - SOC_1) / (SOC_f^2 - SOC_1^2)}.$$ \hspace{1cm} (14)$$

Fig. 4 shows the optimal charging efficiency as a function of initial SOC and $(t_f/RC)$ for the SC cell. Fig. 4 implies the expected result that longer charging times and/or smaller RC values improve the charging efficiency. When $t_f$ approaches infinity, the charging efficiency approaches 1, which is a 100% improvement over the case with CV charging. Another observation is that beginning the charging from a higher initial SOC and also charging within a narrower range of SOC result in improved efficiency. Furthermore, (14) and the first two profiles that are on top of each other in Fig. 4 show that when starting from zero initial SOC, the charging efficiency...
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**III. CHARGING OF THE LEAD-ACID BATTERY**

**A. Lead-Acid Battery Model and Specifications**

The lead-acid battery used in this paper is an AP-12220EV-NB module. The module specifications are listed in Table III.

The real capacity of the module is obtained by discharging the fully charged module with a low current of 0.55 A from the upper voltage limit to the lower voltage limit. This measured capacity is 19.7 Ah. Specifically designed pulse-relaxation tests, such as the method used in [47], is utilized to estimate the total internal resistance $R$ of the cell as a function of $SOC$.

Fig. 5 shows that the internal resistance of lead-acid battery strongly depends on $SOC$.

The OCV of the lead-acid battery is obtained by applying a small current of 0.05 A to charge the battery from zero to full charge. The recorded OCV for this battery as a function of $SOC$ is shown in Fig. 6.

**B. Constant Power Charging of the Lead-Acid Battery**

The relationship between the total internal resistance and $SOC$ of the lead-acid battery is approximated by fitting a second-order polynomial to the profile in Fig. 5 as follows:

$$ R = a_1SOC^2 + a_2SOC + a_3. $$

(15)
TABLE IV

POLYNOMIAL COEFFICIENTS FOR THE OCV AND R AS A FUNCTION OF SOC

<table>
<thead>
<tr>
<th>a1</th>
<th>a2</th>
<th>a3</th>
<th>b1</th>
<th>b2</th>
<th>b3</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.098</td>
<td>-0.12</td>
<td>0.061</td>
<td>-0.56</td>
<td>2.2</td>
<td>11</td>
</tr>
</tbody>
</table>

A second-order polynomial is fitted to the OCV as a function of SOC profile in Fig. 6 as follows:

\[ OCV = b_1 SOC^2 + b_2 SOC + b_3. \] (16)

The polynomial coefficients are listed in Table IV.

As the lead-acid battery is modeled with a total internal resistance of \( R \), applying Kirchhoff’s voltage law to the circuit that is charging the lead-acid battery with a CP source \( (P_0) \) results in

\[ I_{cp} = \frac{-OCV + \sqrt{OCV^2 + 4RP_0}}{2R}. \] (17)

Substituting (15) and (16) in (17) and solving the nonlinear differential equation, the charge and CP charging current are obtained. Consider charging the empty lead-acid battery to full charge in 1 h where \( P_0 = 248 \text{ W} \). The maximum charge storable in this lead-acid battery is 19.7 Ah \( \times 3600 = 70920 \) Coulombs. Fig. 7 shows the charge and CP charging current profiles for charging in 1 h.

For the fast charging case, consider charging the same lead-acid module with a CP of 3660 W, which is equivalent to charging the module in 6 min. Fig. 8 shows that for the lead-acid battery, both the magnitude and shape of the CP charging current vary for the slow and fast charging cases.

C. Optimal Charging of the Lead-Acid Battery

The lead-acid battery is modeled by a single internal resistance in [48], where the only state is the SOC of the battery governed by (7). The objective is to minimize the losses associated with the total internal resistance as in (8). Therefore, the Hamiltonian is

\[ H(x, u, t) = R(x_1)u^2(t) + \dot{\lambda}_2(t)\frac{u(t)}{q_{max}} \] (18)

where \( R \) is the total internal resistance and \( \dot{\lambda}_2(t) \) is the costate. \( R(x_1) \) also shown in Fig. 5 is approximated by the second-order polynomial in (15). The necessary conditions to be satisfied are

\[ -\frac{\partial H}{\partial x_1} = -\frac{dR(x_1)}{dx_1}u^2(t) = \frac{d}{dt}\dot{\lambda}_2(t) \] (19)

\[ \frac{\partial H}{\partial u} = 2R(x_1)u(t) + \frac{\dot{\lambda}_2(t)}{q_{max}} = 0. \] (20)

Solving for \( u(t) \) in (20), the optimal charging current is obtained as follows:

\[ u_{opt}(t) = -\frac{1}{2q_{max}}\frac{1}{R(x_1)}\dot{\lambda}_2(t). \] (21)

Substituting \( u_{opt}(t) \) from (21) in (7) and (19), the consecutive set of two coupled nonlinear ordinary differential equations (ODEs) are obtained

\[ \frac{dx_1(t)}{dt} = -\frac{1}{2q_{max}^2}\frac{1}{R(x_1)}\dot{\lambda}_2(t) \] (22)

\[ \frac{d\dot{\lambda}_2(t)}{dt} = -\frac{1}{4q_{max}^2}\frac{dR(x_1)}{dx_1}\frac{1}{R^2(x_1)}\dot{\lambda}_2^2(t). \] (23)

Charging the lead-acid battery in \( t_f \) units of time from zero to full charge requires the initial and final conditions to be satisfied

\[ x_1(0) = SOCi, \quad x_1(t_f) = SOCf. \] (24)

The system of two nonlinear ODEs with one initial and another final condition forms a TPBVP, which could only be solved using numerical methods. One way to solve this system of ODEs is to specify the initial condition for the SOC and iteratively guess the initial condition for \( \dot{\lambda}_2 \) until SOC reaches the final specified value. Consider the case of charging the lead-acid battery module from zero to full charge in 1 h. Fig. 9 shows the variation of optimal charging current, SOC, and \( \dot{\lambda}_2 \) with time.

As shown in the numerical results, the optimal charging current for lead-acid battery, unlike the SC, is not constant. In order to compare the CC charging with the optimal charging strategy, the energy losses in both methods are calculated. For the case of charging the lead-acid battery from zero to full charge in 1 h, the resistive losses in the optimal charging strategy are 46.18 kJ compared with 48.9 kJ for CC charging.
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PA RV IN I et al.: HEURISTIC VERSUS OPTIMAL CHARGING OF SCs, Li-Ion, AND LEAD-ACID BATTERIES

Fig. 9. Optimal charging current, \( \text{SOC} \), and \( \lambda \) profiles for charging the lead-acid battery from zero to full charge in 1 h.

**TABLE V**

<table>
<thead>
<tr>
<th></th>
<th>Slow Charging (1 h)</th>
<th>Fast Charging (6 min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CC</td>
<td>95.19</td>
<td>64.43</td>
</tr>
<tr>
<td>CP</td>
<td>95.34</td>
<td>67.46</td>
</tr>
<tr>
<td>Optimal</td>
<td>95.44</td>
<td>67.70</td>
</tr>
</tbody>
</table>

This is a 5.5% of less energy converted to heat which could be significant in thermal management of battery packs.

**D. Efficiency Analysis for the Lead-Acid Battery**

The efficiency for three charging strategies including CP, CC, and optimal charging is calculated based on the definition of efficiency

\[
\rho = \frac{E_{\text{battery}}}{E_{\text{battery}} + E_{\text{loss}}}. \quad (25)
\]

According to the lead-acid battery specification listed in Table III, \( E_{\text{battery}} \) is 268.8 Wh for this module. The energy loss (\( E_{\text{loss}} \)) is obtained by numerically integrating the power loss (\( RI^2 \)) during the charging time. Table V compares the efficiency values for charging the lead-acid battery from zero to full charge for three strategies and two charging times. The efficiency of optimal charging is slightly higher than the other two strategies as expected. If factors, such as cost of supplying a non-CC are considered, one may prefer to charge the lead-acid battery with CC and neglect the effect of the slightly lower charging efficiency. The efficiency of CP charging is higher than CC charging for the lead-acid battery.

**IV. CHARGING OF THE LI-ION BATTERY**

**A. Li-Ion Battery Model and Specifications**

The Li-ion battery used in this paper is an A123-26650 cell with LiFePO\(_4\) chemistry. The cell specifications are listed in Table VI. The cell model is developed using pulse-relaxation experiments to identify equivalent electric circuit model parameters. The estimation is performed by minimizing the least square error between the experimental and modeled terminal voltages [49].

For the Li-ion battery, two models are studied. First, only electronic resistance \( R_s \) is considered, and in the next step, the effect of polarization resistance \( R_1 \) is also included by adding a single \( RC \) branch to the model as shown in Fig. 10. The dependence of model parameters \( R_s, R_1, \) and \( C_1 \) on \( SOC \) can be neglected at room temperature [49]. The values for \( R_s, R_1, \) and \( C_1 \) are \( 0.01 \Omega, 0.016 \Omega, \) and \( 2200 \text{F} \), respectively.

The \( OCV \) of the Li-ion battery as a function of \( SOC \) is shown in Fig. 11. As shown in Fig. 11, the \( OCV \) can be approximated by a linear function using the \( OCV \) data in the range of 10%–95% \( SOC \). This linear fit makes the analytical efficiency analysis possible. The linear approximation is governed by

\[
OCV(t) = aSOC(t) + b. \quad (26)
\]

The relationship between \( OCV(t) \) and the charge stored \( q(t) \) in the battery is obtained by substituting the definition of
Solving for the CP charging current for the Li-ion battery. First scenario: in this step, only \( I_1 \) and \( I_2 \) are considered. Therefore, the cost function to be minimized is

\[
J_2 = \int_0^t \left[ R_s u_2^2(t) + R_1 x_2^2(t) \right] dt.
\]

The Hamiltonian, in this case, is given by the subsequent equation

\[
H(x, u, t) = R_s u(t) + R_1 x_2^2(t) + \lambda_3 \frac{u(t)}{q_{\text{max}}} + \lambda_4 \frac{u(t) - x_2(t)}{R_1 C_1}
\]

where \( \lambda_3 \) and \( \lambda_4 \) are the costates. The necessary conditions for optimality are

\[
-\frac{\partial H}{\partial x_1} = \frac{d}{dt} \lambda_3, \quad -\frac{\partial H}{\partial x_2} = \frac{d}{dt} \lambda_4, \quad \frac{\partial H}{\partial u} = 0.
\]

From the first two conditions in (35), the dynamics of the costates is derived and the solution to the third condition is the optimal input as follows:

\[
u_{\text{opt}}(t) = \left[ -\frac{1}{2 R_s q_{\text{max}}} \right] \lambda_3(t) + \left[ -\frac{1}{2} R_s R_1 C_1 \right] \lambda_4(t).
\]
Substituting the optimal input into the state equations of (32), the optimal state dynamics is derived. The result is a set of four linear first-order ODEs

\[
\begin{align*}
\frac{d}{dt}x_1(t) &= a_1 \dot{\lambda}_3(t) + a_2 \dot{\lambda}_4(t) \\
\frac{d}{dt}x_2(t) &= b_1 x_2(t) + b_2 \dot{\lambda}_3(t) + b_3 \dot{\lambda}_4(t) \\
\frac{d}{dt}x_3(t) &= 0 \\
\frac{d}{dt}x_4(t) &= c_1 x_2(t) + c_2 \dot{\lambda}_4(t)
\end{align*}
\]  

(37)

where \(a_1, a_2, b_1, b_2, b_3, c_1,\) and \(c_2\) are constant parameters equal to

\[
\begin{align*}
a_1 &= -\frac{1}{2R_\text{s}q_{\text{max}}^2}, \quad a_2 = -\frac{1}{2R_\text{s}R_1C_1q_{\text{max}}}, \\
b_1 &= -\frac{1}{R_1C_1}, \quad b_2 = -\frac{1}{2R_\text{s}R_1C_1q_{\text{max}}}, \\
b_3 &= -\frac{1}{2R_\text{s}R_1^2C_1}, \quad c_1 = -2R_1, \quad c_2 = \frac{1}{R_1C_1}.
\end{align*}
\]  

(38)

Solving this system of coupled linear ODEs simultaneously results in four algebraic equations with four unknowns. The unknown constants are obtained by applying the boundary conditions specific to this problem, which consist of two initial and two final conditions. The initial and final conditions for \(x_1 = \text{SOC}\) are similar to (24). On the other hand, in this specific problem, the charging time is specified and fixed while the values of the second state at the initial and final time are free. This results in the succeeding equations for the remaining two boundary conditions [46]

\[
\begin{align*}
\frac{\partial h}{\partial x_2}(x_2(t_0)) &= \dot{\lambda}_4(t_0) = 0 & \text{Initial condition for } x_2 \\
\frac{\partial h}{\partial x_2}(x_2(t_f)) &= \dot{\lambda}_4(t_f) = 0 & \text{Final condition for } x_2.
\end{align*}
\]  

(39)

In general, \(h(x(t_f), t_f)\) is the term involving the final states and final time in the cost function, which, in this paper, is zero. Given all boundary conditions, one can solve for the states and costates, and thus, the optimal input is obtained. Consider charging a battery cell from zero charge \(\text{SOC}_1 = x_1(0) = 0\) to full charge \(\text{SOC}_1 = x_1(t_f) = 1\) in 1 h. The result for this example is shown in Fig. 13.

The optimal charging current for this scenario is slightly different from the result of the first scenario. The optimal input in this case is almost a CC equal to 2.5 A, in the majority of times. It may be insightful to also show the result for a fast charging case. Fig. 14 shows the optimal charging current and the two states of the system when the cell is charged from zero to full charge in 6 min.

This charging strategy may not be practical due to thermal and physical constraints plus safety and lifetime issues. However, it may be interesting to observe that by reducing the charging time, the optimal profile differs from the CC result observed in the first scenario and also long charging times in the second scenario.

**D. Efficiency Analysis for the Li-Ion Battery**

In order to find the optimal charging efficiency, the total energy stored in the battery and energy loss is required. Assuming a constant total internal resistance \(R = R_\text{s} + R_1 = 0.026 \, \Omega\) makes the analytical efficiency analysis possible. The energy loss in the battery during optimal charging is already known and is equal to \(\int_0^{t_f} Ru_{\text{opt}}^2 dt\). The total energy stored in the battery is

\[
E_{\text{battery}} = \int_0^{t_f} V I dt = \int_0^{t_f} V \frac{dq(t)}{dt} dt = \int_0^{q_{\text{max}}} V dq \quad (40)
\]
where \( I, V, \) and \( q \) are the battery current, \( OCV, \) and the charge in Ah, respectively. Using the linear relationship between \( OCV \) and the charge stored in the battery from (27) and the definition of \( SOC, \) the maximum energy stored in the battery is obtained as follows:

\[
E_{\text{battery}} = q_{\text{max}} (SOC_f - SOC_i) \left[ a (SOC_i + SOC_f) + b \right] 
\]

(41)

where the unit for energy is watt-hour (Wh). The real maximum amount of energy which the battery can store is obtained by integrating the original \( (OCV - q) \) profile, which results in 8.2 Wh for the cell used in this paper. Using (41) and charging the battery from 0% to 100%, the maximum battery energy calculated is 8.26 Wh. This illustrates that the linear approximation of \( OCV \) for Li-ion battery is an effective approach to perform analytical efficiency analysis. Substituting the expressions for \( E_{\text{loss}} \) and \( E_{\text{battery}} \) in (25), the optimal charging efficiency for Li-ion battery is obtained as follows:

\[
\rho_{\text{opt}} = \frac{1}{1 + \frac{R q_{\text{max}} (SOC_f - SOC_i)}{t_f (\frac{1}{2} a (SOC_i + SOC_f) + b)}} 
\]

(42)

where \( t_f \) is the charging time in hours and \( R \) is the total internal resistance in ohms. Similar to the SC, starting the charging from a higher initial \( SOC \) results in better efficiency. Also faster charging will result in higher currents and lower efficiency. For the Li-ion battery, the CP and the two optimal charging scenarios are almost identical in terms of the charging current profiles and also the efficiency values.

V. EFFECT OF TEMPERATURE ON OPTIMAL CHARGING OF THE LI-ION BATTERY

In this section, unlike the unconstrained cases solved in Sections II–IV, the optimal problem is solved subject to voltage and temperature constraints. This approach represents a more realistic solution of the optimal charging of the Li-ion battery.

A. Electrothermal Model of the Li-Ion Battery

In order to investigate the effect of temperature on the optimal charging current, a thermal model needs to be coupled with the electrical model and integrated in the optimal charging formulation. The electrical model considered for this section is a single resistance model, which represents the total internal resistance as the sum of the electronic and polarization resistances \((R = R_e + R_i)\). The dynamics of the electrical model is governed by the single state equation (7) with current being the input and \( SOC \) as the state. On the other hand, the thermal model is a reduced order model represented by two states. For further details on reducing the governing PDE to two linear ODEs, please refer to [51]. The thermal model is identified and validated for the A123-26650 cell with specifications listed in Table VI. The state space representation of the thermal model is

\[
\dot{x} = Ax + Bu, \quad y = Cx + Du 
\]

(43)

\[
\begin{align*}
A &= \begin{bmatrix}
-48\beta h & -15\beta h \\
24k_i + rh & 120\beta (4k_i + rh)
\end{bmatrix} \\
B &= \begin{bmatrix}
\beta & 48\beta h \\
24k_i + rh & 320\beta h
\end{bmatrix} \\
C &= \begin{bmatrix}
24k_i + rh & 8(24k_i + rh) \\
48k_i + 2rh & 15k_i
\end{bmatrix} \\
D &= \begin{bmatrix}
0 & 24k_i + rh \\
0 & 24k_i + rh
\end{bmatrix}
\end{align*}
\]

where \( x = [\bar{T} \bar{y}]^\top, \ u = [QT_\infty]^\top, \) and \( y = [T_c T_s]^\top \) are state, input, and output vectors, respectively. The states of the thermal model are the volume-averaged temperature \( \bar{T} \) in Kelvin (K) and the volume-averaged temperature gradient \( \bar{y} \) in (K/m). The inputs are the ambient temperature \( T_\infty \) in Kelvin and the total heat generation rate \( Q. \) The outputs of the model are the battery’s surface temperature \( T_c \) and core temperature \( T_s \) both in Kelvin. The linear system matrices \( A-D \) are

\[
\begin{align*}
\text{Table VII} \\
\text{LI-ION PHYSICAL AND THERMAL PARAMETERS}
\end{align*}
\]

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radius</td>
<td>( r )</td>
<td>12.93e-3</td>
<td>m</td>
</tr>
<tr>
<td>Volume</td>
<td>( V_{\text{cell}} )</td>
<td>3.4219e-5</td>
<td>m³</td>
</tr>
<tr>
<td>Density</td>
<td>( \rho )</td>
<td>2047</td>
<td>( \frac{\text{g}}{\text{cm}^3} )</td>
</tr>
<tr>
<td>Specific Heat</td>
<td>( c_p )</td>
<td>1109.2</td>
<td>( \frac{\text{J}}{\text{g} \cdot \text{K}} )</td>
</tr>
<tr>
<td>Thermal Conductivity</td>
<td>( k_1 )</td>
<td>0.61</td>
<td>( \frac{\text{W}}{\text{m} \cdot \text{K}} )</td>
</tr>
<tr>
<td>Convection Coefficient</td>
<td>( h )</td>
<td>58.6</td>
<td>( \frac{\text{W}}{\text{cm}^2 \cdot \text{K}} )</td>
</tr>
</tbody>
</table>

where \( x = [\bar{T} \bar{y}]^\top, u = [QT_\infty]^\top, \) and \( y = [T_c T_s]^\top \) are state, input, and output vectors, respectively. The states of the thermal model are the volume-averaged temperature \( \bar{T} \) in Kelvin (K) and the volume-averaged temperature gradient \( \bar{y} \) in (K/m). The inputs are the ambient temperature \( T_\infty \) in Kelvin and the total heat generation rate \( Q. \) The outputs of the model are the battery’s surface temperature \( T_c \) and core temperature \( T_s \) both in Kelvin. The linear system matrices \( A-D \) are
Fig. 15. Internal resistance $R$ as a function of core temperature for the Li-ion battery.

![Fig. 15. Internal resistance $R$ as a function of core temperature for the Li-ion battery.](image)

**TABLE VIII**

<table>
<thead>
<tr>
<th>$c_1$</th>
<th>$c_2$</th>
<th>$c_3$</th>
<th>$c_4$</th>
<th>$c_5$</th>
<th>$c_6$</th>
<th>$c_7$</th>
<th>$d_1$</th>
<th>$d_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0063599</td>
<td>-0.01179</td>
<td>-0.001717</td>
<td>0.0071517</td>
<td>0.0094349</td>
<td>-0.023463</td>
<td>0.035803</td>
<td>-284.77</td>
<td>22.165</td>
</tr>
</tbody>
</table>

**B. Constrained Optimal Fast Charging of the Li-Ion Battery**

The described electrothermal model consists of three states. Formulating the efficiency maximization problem using PMP will result in three state equations and three costate dynamic equations with three unknown initial conditions that need to be guessed. This makes the problem tedious to solve using normal nonlinear ODE solvers. DP is used instead to solve this optimal control problem relying on Bellman’s principal of optimality. We focus on the optimal charging of the Li-ion battery by realizing that the total internal resistance is a function of core temperature of the cell. The objective similar to the preceding scenarios is to minimize the resistive losses associated with the total internal resistance

$$J = \int_0^{t_f} R(T_c)u^2(t)dt$$  \hspace{1cm} (45)

where $T_c$ is the core temperature. Furthermore, we are interested in fast charging while considering the effect of voltage and temperature constraints. For this reason, the charging time of $t_f = 10$ min is chosen. The initial charging temperature is $T_{\infty} = 25$ °C. The resolution for the time in the DP code is 20 s. At each time instant $t_i$, all three states of the electrothermal model are quantized and represented by $(SOC_{i}, T_{i}, \gamma_i)$. Fig. 16 demonstrates the implementation of DP.

![Fig. 16. Illustration of the DP grid and sample transitions.](image)

**TABLE IX**

<table>
<thead>
<tr>
<th>Unconstrained CC and Optimal Charging in 10 min</th>
<th>Unconstrained</th>
<th>DP</th>
</tr>
</thead>
<tbody>
<tr>
<td>Charging Current (A)</td>
<td>15</td>
<td>20 max.</td>
</tr>
<tr>
<td>Core Temperature (°C)</td>
<td>43 max.</td>
<td>42 max.</td>
</tr>
<tr>
<td>Terminal Voltage (V)</td>
<td>3.8 max.</td>
<td>3.7 max.</td>
</tr>
<tr>
<td>State-of-Charge @ $t_f$</td>
<td>100%</td>
<td>100%</td>
</tr>
<tr>
<td>Energy Loss (J)</td>
<td>2682</td>
<td>2638</td>
</tr>
<tr>
<td>Efficiency</td>
<td>91.72%</td>
<td>91.84%</td>
</tr>
</tbody>
</table>

**CC**: Constant Current  
**DP**: Dynamic Programming

states, the optimal control input, which in this paper is the optimal charging current, is obtained.

Three fast charging scenarios are considered as follows.

1) **Unconstrained Charging**: Similar to the other sections in this paper, the constraints are relaxed and the efficiencies obtained from DP and CC charging are compared for charging from zero to full charge in 10 min. The results are summarized in Table IX where the optimal charging efficiency is slightly higher than CC charging. To be consistent and in order to compare the results with the constrained scenarios, the unconstrained CC and DP charging scenario is resimulated for charging from 0% to 90% which the results are given in the first column of Table X, indicated as CC and DP1, respectively.

2) **Voltage-Constrained Charging**: A real-world implementation would require constraints on the terminal voltage and core temperature. In the second case, the similar optimal charging problem is solved subject to a voltage constraint given in (46) for both DP and CC charging. In this scenario, the battery is charged from 0% to 90%, since fully charging is not possible due to the voltage constraint as well as the short charging time of 10 min. The results are given in the second column of Table X, indicated as CCCV and DP2 where CCCV charging is actually the CC charging with the voltage constraint imposed.
3) Double Voltage/Temperature Constrained Charging: In the final scenario, keeping the terminal voltage constraint, the optimal trajectory and efficiency are obtained by adding the core temperature constraint of (47) to both the DP and CC charging methods. Again, the battery is charged from 0% to 90%; and the results are given in the third column of Table X, indicated as CCCV$^*$ and DP3 where CCCV$^*$ is the notation for CC charging with both voltage and temperature constraints active.

$$2V \leq V_T \leq 3.6V$$

(46)

$$T_c \leq 39^\circ C$$

(47)

Neither CC nor DP directly apply a limit on the maximum charging current as the voltage and temperature constraints indirectly result in limited charging current when they become active. The voltage and temperature limits in (46) and (47) are chosen based on the manufacturer’s recommendation.

Fig. 17 and the odd columns of Table X present the results for the CC charging. During CCCV charging, as the inset in the first subplot of Fig. 17 shows, the current drops at the end of charging as the terminal voltage hits the limit of 3.6 V. During the CCCV$^*$ charging, the amount of initial current (20 A) is chosen, such that the battery could be charged to 90% SOC. According to Fig. 17, at about 30 and 200 s into the CCCV$^*$ charging, the voltage and temperature limits are reached, respectively, which results in a stepwise decrease in current. Comparing the efficiencies listed in Table X for CC, CCCV, and CCCV$^*$ shows that imposing constraints on the CC charging results in smaller values for efficiency.

Fig. 18 and the even columns of Table X show the results for the optimal charging using DP. As it was mentioned earlier, the unconstrained DP, voltage-constrained, and the double voltage/temperature-constrained problems are indicated as DP1, DP2, and DP3, respectively. The optimal charging has a slightly higher efficiency compared with the CC charging scenarios. The general trajectory of all the three DP cases shows a warm-up period at the beginning of charging followed by a CC charging section. The physical reason behind this behavior is that the higher current at the beginning of charging results in a rapid increase in core temperature, which consequently decreases the total internal resistance. The lower total internal resistance is in favor of minimizing the charging losses according to the objective function in (45). The larger current at the beginning of charging results in terminal voltage reaching the upper limit, which results in a decrease in current for DP2 and DP3 scenarios. For DP3, as also shown via the inset in the second subplot in Fig. 18, the temperature limit is activated toward the end of charging, which results in an instant decrease in current to avoid overheating. The DP results are based on a 20 s resolution on time which maybe the reason for the ripples in current specifically obvious in the DP2 case.

These optimal solutions are computed using Clemson University’s Palmetto cluster to facilitate the high memory requirements of the DP implementation. In this particular problem, the three state variables $SOC$, $\overline{T}$, and $\overline{\gamma}$ are quantized to $n_{SOC}$ = 200, $n_\overline{T}$ = 25, and $n_\overline{\gamma}$ = 25, respectively. This requires each grid of Fig. 16 to accommodate $n_{SOC} \times n_\overline{T} \times n_\overline{\gamma} = 125 000$ cells. Moreover, each grid is stored by 125 000 x 125 000 cells in the implementation as each cell at time instant $t_{i-1}$ has also 125 000 possible transitions to the next cell at time instant $t_i$. In the implemented DP code, ten variables are involved in the backward DP computation with mixed single/double precision (6 Byte on average). This would mean that the minimum required memory allocation is $10 \times 125 000 \times 125 000 \times 6$ Byte or $\approx$ 870 GB. The involved variables are the two inputs to the objective function ($R(T_c)$ and $u(t)$), three state variables ($SOC$, $\overline{T}$, $\overline{\gamma}$), one combined constraint based on (46) and (47), and four cost functions.
These cost functions include one unconstrained cost-to-go function at \( t_i \) and two constrained cost-to-go functions at \( t_i \) and \( t_j \) intervals.

VI. CONCLUSION

This paper solved the optimal charging problem for different energy storage systems considering different levels of model complexity and also compared the optimal charging current with CP, CC, and CV charging strategies. Efficiency analysis was also performed to compare different charging strategies. For the SC and the first scenario of the Li-ion battery, the optimal charging strategy is CC. However, for the Si-CN electrode, the optimal charging strategy is a combination of CC and CV. In the Li-ion case, there is no much difference in applying CP or CC considering the efficiency and shape of the current profile. For the lead-acid battery, the optimal strategy has a better efficiency than CP charging, while CC charging has the least efficiency. Finally, the effect of temperature dependent model parameters as well as the voltage and temperature constraints on the optimal fast charging of the Li-ion battery was investigated. This constrained optimal control problem was solved using DP and was compared with CC charging. The results show that the charging efficiency of the optimal scenario is slightly higher than CC charging.

REFERENCES


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