

State of Charge (SOC) Governed Fast Charging Method for Lithium Based Batteries*

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Abstract: The State of Charge (SOC) governed fast charging method for secondary lithium based batteries proposed herein charges a battery many times faster than the normal Constant Current-Constant Voltage (CC-CV) charging and reduces the side-effects generally accompanied by various fast charging methods. The proposed charging algorithm takes into account the varying internal impedance of the battery at different SOC levels and minimizes voltage fluctuations. The charging method has been demonstrated using a simulated lab test on a 3.72V lithium-ion battery cell from Navitasys, LLC and modeled using the Battery & Fuel cells module along with the Events interface of COMSOL Multiphysics 4.3b. The experimental and modeling results were compared with other state-of-the art fast charging methods available in the literature.

Keywords: Fast charging, lithium batteries, State of Charge, COMSOL Multiphysics

1. Introduction

Considering the world's current energy starved scenario and the requirement to persistently met the stringent emissions norms, electric vehicles (EVs) and hybrid-electric vehicles (HEVs) will soon become our most common modes of transportation. The numbers of EVs and HEVs sold worldwide are projected to increase by 3 times in the year 2020. Almost 100% of the EV and PHEVs and 50-60% of the HEVs will be powered by lithium batteries as amongst the available battery technologies, lithium batteries have better automotive advantages which include higher gravimetric energy density, higher cyclability, charge efficiency without memory effect, etc. However, its cost, safety issues and long charging durations are the major concerns.

CC-CV charging is the most widely accepted and universally used charging profile for lithium chemistries as it not only provides safest charging but also most optimum battery performance and maximum battery life. It takes ~ 1.5-2 hours to completely charge a battery in CC-CV mode and when we see EVs and HEVs as our soon to be modes of transportation and when there is a constant comparison between the time it takes to fill a gasoline tank of a vehicle to the time required for charging a battery, the feeling is far from comforting. Hence, there is a lot of initiative for the development of fast charging methods and infrastructures worldwide. As a result, large numbers of fast charging methods [1-5] have been devised in the past decade; but most of them negatively impacts the battery performance in the long run and reduces its cycle-life due to pumping of very high currents into the battery within very short time durations. This leads to irreversible capacity loss as a result of dendrite formation which further results in internal short-circuits and hence permanent battery failure. Yury M. Podrazhansky et.al [6] discloses a method and an apparatus to carry out rapid charging of lithium based batteries while eliminating performance degradation as a result of formation of resistive layers on electrode surfaces, dendritic formation and electrolyte decomposition.

In the same line of work, a novel fast-charging method (Figure 1) is presented in this paper for secondary lithium based batteries that is suitable for charging any lithium-ion, lithium-polymer and solid-state lithium chemistry. The charging profile has been carefully constructed by taking close consideration of the physical processes involved in the battery during charging and the varying impedance of the battery at different SOC levels so as to obtain optimum battery performance while charging it rapidly and at the same time to avoid internal stress within the battery active material and at the

electrode | electrolyte interfaces so as to obtain maximum cycle life.

2. The proposed fast charging method

Charging and discharging process in lithium based batteries primarily involves the shuttling of lithium-ions between the electrodes and lithium-ion intercalation in the interstices of the cathode and anode respectively. Since the time-constant for solid-state diffusion within the electrodes is much higher than the time constant for diffusion in liquids, it is critical to provide enough settling time for the lithium-ions to move into the vacant sites of the host electrodes. Otherwise, unmindful fast charging may result in dendrite formation or lithium plating as a result of accumulation of lithium-ions on the anode surface and apart from capacity loss and cycle-life degradation, it can pose serious safety threats in the form of short-circuit, overheating leading to cell bulging and also explosion in extreme cases.

It is advisable to start the charging process from a lower current rate if the state of the battery is unknown. Also, the battery encounters high internal impedance at lower SOC levels which may be the result of deep-discharge. Impedance is also contributed as a result of prolonged storage duration and storage at lower temperatures. To account for higher impedance at lower SOC levels, the first charging stage, C_1 consists of a multistage Constant Current region ($m\text{-CC}_i$) where gradually increasing current pulses of amplitude varying from $0.5C - 2C$ are applied to the battery based on its SOC with alternate rest periods. This stage extends from $0 = \text{SOC} < 50$.

The second charging stage, C_2 ($50 \leq \text{SOC} < 80$) consists of a multistage Constant Current – Constant Voltage ($m\text{-CC-CV}$) region. High current pulses of amplitude in the range of $2C - 8C$ are applied to the battery in the CC step as lithium batteries exhibits higher charge acceptance and a flat discharge plateau in this region. A CV step is applied after each high charging current pulse, which reduces the voltage fluctuations by restricting the battery voltage from dropping drastically after the CC step. The CV step allows more time for the Li^+ ions to diffuse into the interstices of the anode

thereby smoothening the concentration gradient on the anode surface. This step helps in eliminating the chances of lithium plating on anode surface.

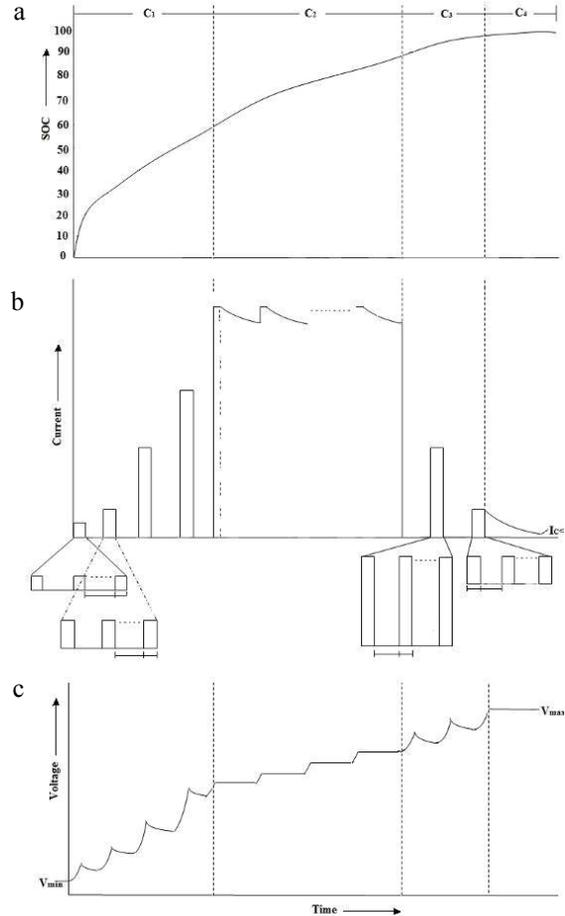


Figure 1. The proposed fast charging method showing the (a) SOC (b) Current and (c) Voltage profiles

The third charging stage, C_3 consists of another multistage Constant Current region ($m\text{-CC}_r$) where gradually decreasing current pulses of amplitude varying from $0.5C - 2C$ are applied to the battery based on its SOC with alternate rest periods in order to account for the lower charge acceptance by the battery at higher SOC levels ($80 \leq \text{SOC} < 95$).

The final charging stage, C_4 consists of a single CV step which is applied to the battery at V_{max} (charge cut-off voltage) after it has attained $\geq 95\%$ SOC. The gradually decreasing current in

this stage slowly brings the battery to equilibrium at the end of the charge. Charging is terminated once the charging current drops to about 0.1C - 0.05C

The charging profile may or may not follow the stages in the exact order as proposed herein depending upon the SOC level of the battery prior to charging.

3. Use of COMSOL Multiphysics

A lithium-ion cell of lithium-manganese oxide, LiMn_2O_4 chemistry has been modeled using the Batteries & Fuel cells module and the proposed charging profile has been simulated using the Events interface of COMSOL Multiphysics 4.3b.

The 1D lithium-ion battery model consists of the following five domains (Figure 2):

- Negative current collector (Copper) of length L_{neg_cc}
- Negative electrode (Li_xC_6) of length L_{neg}
- Separator of length L_{sep} with electrolyte (1:1 EC:DEC in LiPF_6 salt)
- Positive electrode ($\text{Li}_x\text{Mn}_2\text{O}_4$) of length L_{pos}
- Positive current collector (Aluminum) of length L_{pos_cc}

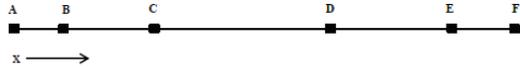


Figure 2. 1D model of the lithium-ion battery where A is $x=0$, B is $x=L_{neg_cc}$, C is $x=L_{neg_cc}+L_{neg}$, D is $x=L_{neg_cc}+L_{neg}+L_{sep}$, E is $x=L_{neg_cc}+L_{neg}+L_{sep}+L_{pos}$ and F is $x=L_{neg_cc}+L_{neg}+L_{sep}+L_{pos}+L_{pos_cc}$

The physical and electrical properties of all the domain materials have been derived from the Material Library of COMSOL Multiphysics.

The electrode reactions have been modeled using the Butler-Volmer kinetic equation which is given by the expression:

$$J_n = \frac{i_0}{F} \left[\exp\left(\frac{\alpha_a F \eta}{RT}\right) - \exp\left(\frac{-\alpha_c F \eta}{RT}\right) \right]$$

where, J_n is the pore wall flux, η is the over-potential due to electrode reaction, i_0 is the exchange current density and α_a and α_c are the symmetry factor for the negative and the positive electrodes respectively.

Charge balance in the electrolyte is governed by the liquid phase Ohm's law given by:

$$i_l = -\sigma_{l,eff} \nabla \phi_l + \frac{2RT\sigma_{l,eff}}{F} \left(1 + \frac{\partial \ln f}{\partial \ln c_l} \right) (1 - t_+^0) \nabla \ln c_l$$

where, $\sigma_{l,eff}$ is the effective conductivity of the ions present in the electrolyte and ϕ_l is the liquid phase potential.

Charge balance inside the electrodes is modeled using the solid phase Ohm's law:

$$i_s = -\sigma_{s,eff} \nabla \phi_s$$

where, $\sigma_{s,eff}$ is the effective conductivity of the ions in the solid phase and ϕ_s is the solid phase potential.

Diffusion or mass-transfer in the electrolyte has been modeled using the Fick's second law, given by:

$$\epsilon_l \frac{\partial c_l}{\partial t} = \frac{\partial}{\partial x} \left(D_{l,eff} \frac{\partial c_l}{\partial x} \right) + (1 - t_+^0) a_s J_n$$

where, c_l is the salt concentration in the solution phase, $D_{l,eff}$ is the effective diffusion co-efficient, a_s is the surface area of the electrode per unit volume and t_+^0 is the transference no. of Li^+ ions.

The active materials of the electrodes are assumed to be spherical particles of known radius r_p and the mass transfer within the electrodes is governed by the Fick's second law in spherical co-ordinates.

$$\frac{\partial c_s}{\partial t} = D_s \left[\frac{\partial^2 c_s}{\partial r^2} + \frac{2}{r} \left(\frac{\partial c_s}{\partial r} \right) \right]$$

where, c_s is the salt concentration in the solid phase and D_s is the diffusion co-efficient of Li^+ in the solid phase.

At the surface of the particles, flux is equal to the rate of accumulation or generation of Li^+ due to the electrochemical reaction. It is given by:

$$\frac{\partial c_s}{\partial r} \Big|_{r=r_p} = J_n$$

At the center of the particle, there is no flux.

$$\frac{\partial c_s}{\partial r} \Big|_{r=0} = 0$$

Also, there is no mass-flux at the electrode | current-collector interfaces. At the negative electrode| negative current-collector interface:

$$\frac{\partial c_{s,n}}{\partial x} \Big|_{x=L_{neg_cc}} = 0$$

At the positive electrode| positive current-collector interface:

$$\frac{\partial c_{s,n}}{\partial x} \Big|_{x=L_{neg_cc}+L_{neg}+L_{sep}+L_{pos}} = 0$$

where, $c_{s,p}$ and $c_{s,n}$ are the Li^+ concentrations in the negative and the positive electrodes respectively.

The negative electrode is grounded at $x=0$;

$$\phi_s \Big|_{x=0} = 0$$

A predefined charging profile, i_{app} is applied to the positive electrode;

$$\phi_s \Big|_{x=L_{neg_cc}+L_{neg}+L_{sep}+L_{pos}+L_{pos_cc}} = -i_{app}$$

Double layer capacitance developed at the electrode | electrolyte interfaces as a result of accumulation of ions on the electrode surface is modeled using the expression:

$$i_{dl} = \left(\frac{\partial \phi_s}{\partial t} - \frac{\partial \phi_l}{\partial t} \right) a_{dl} C_{dl}$$

where, a_{dl} is the double layer area.

C_1 and C_3 stages of the charging profile has been generated using a series of explicit events whereas C_2 and C_4 stages has been generated

using a series of implicit events in the Events interface present under the Mathematics node in the Physics tree . An explicit event occurs at predetermined times and can be repeatedly invoked until the desired condition is fulfilled. On the other hand, an implicit event occurs when a condition involving an indicator state is fulfilled. The switching of the charging profile from one stage to another is governed by the SOC of the battery which is given by:

$$SOC = \frac{\int c_s dS}{c_{s,max}L}$$

where, L is the length of the respective electrode.

Further, the applied current is defined using a global ODEs and DAEs interface.

4. Results and discussion

The lithium-ion cell modeled herein has a rated capacity of 10Ah, charge termination voltage, V_{max} of 4.2V, corresponding to 100% SOC and a discharge cut-off voltage of 3V, corresponding to 0% SOC.

The proposed charging profile simulated using COMSOL (Figure 3) consists of 3 charging levels in the C_1 stage (m-CC_i). In the first level of charging from $0.2 = SOC < 0.3$, a constant current of 0.7C (i.e., 7A) is applied to the battery whereas in the second level of charging from $0.3 \leq SOC < 0.4$, constant current of 1C (i.e., 10A) and in the third level of charging from $0.4 \leq SOC < 0.5$, constant current of 1.3C (i.e., 13A) is applied to the battery. In all the levels, current pulse is applied for a period of 3s with alternate rest periods of 6s. In the C_2 charging stage (m-(CC-CV)); six CC-CV steps are performed till the battery SOC reaches 80%. Constant current pulses of 2C (i.e., 20A) is applied in the CC stage followed by a CV step. C_3 charging stage (m-CC_r) is carried out in 3 charging levels. In the first level of charging from $0.80 = SOC < 0.85$, constant current of 1.5C (i.e., 15A) is applied to the battery whereas in the second level of charging from $0.85 \leq SOC < 0.9$, constant current of 1C (i.e., 10A) and in the third level of charging from $0.9 \leq SOC < 0.95$, constant current of 0.8C (i.e., 8A) is applied to the battery for a period of 6s with

alternate rest periods of 3s. Finally, in the C_4 charging stage, constant voltage charging is performed at 4.2V (V_{max}) to attain 100% SOC. The total charging time taken to reach from 0% SOC to 100% SOC using this method is 2500s (~42 minutes).

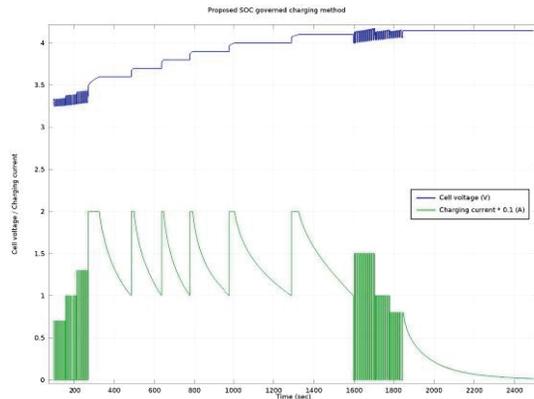


Figure 3. The proposed fast charging method simulated using COMSOL Multiphysics 4.3b showing the cell voltage and charging current profiles

The proposed charging method has been compared with the conventional Constant-Current Constant-Voltage (CC-CV) charging, multistage Constant-Current Constant-Voltage m-(CC-CV) charging [1] and boost charging [5].

In CC-CV charging (Figure 4a), a constant current of $0.5C$ (5A) is applied to the battery in the Constant Current (CC) step till it reaches the charge termination voltage, V_{max} (4.2V). Next, Constant Voltage (CV) charging is performed at V_{max} till the charging current drops to $0.01C$ (0.1A). The charging time taken in this method is 8000s (~2.2 hrs).

The multistage Constant-Current Constant-Voltage m-(CC-CV) charging profile (Figure 4b) consists of two CC-CV stages, i.e., CC-CV-CC-CV. Charging is initiated with a constant current of $2C$ (20A) until the battery voltage reaches its first stage termination voltage, V_1 corresponding to 80% SOC (4V). Next, a constant voltage profile is applied at V_1 (4V) till the charging current drops down to $0.7C$ (7A). The second CC stage follows at a constant charging current of $0.7C$ till the battery attains the charge termination voltage, V_{max} . Finally, a CV step is applied at V_{max} till the charging current drops to

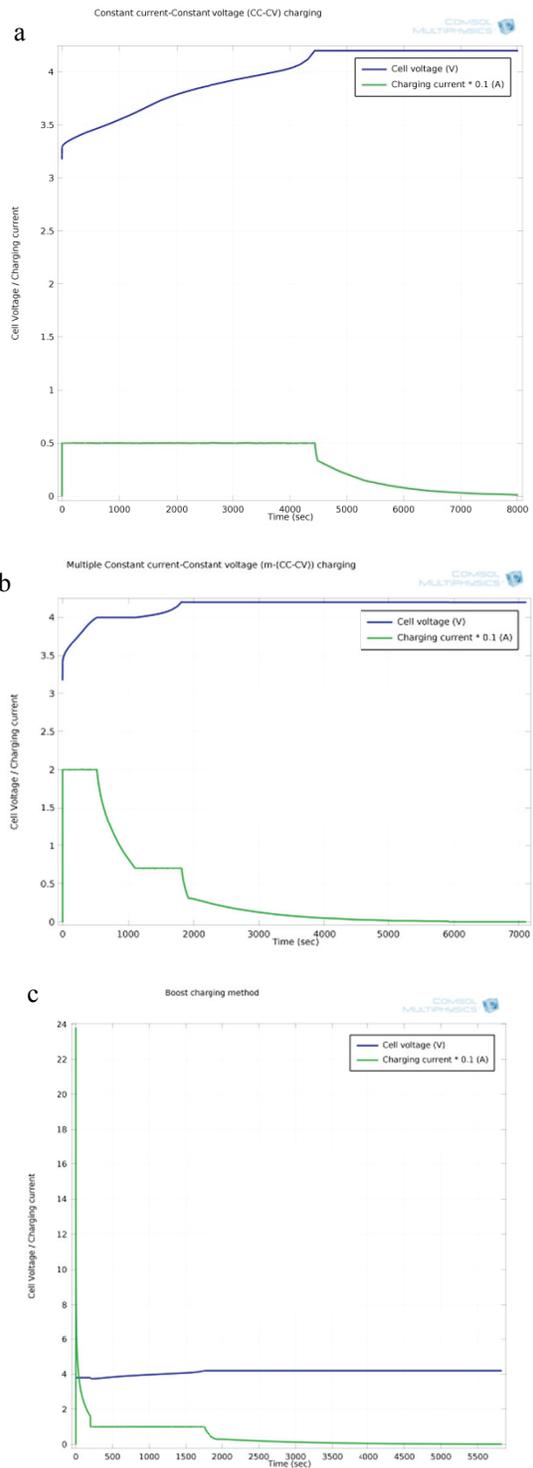


Figure 4. (a) Constant Current- Constant Voltage (CC-CV) charging profile, (b) Multiple Constant Current- Constant Voltage (CC-CV-CC-CV) charging profile, (c) Boost charging profile (CV-CC-CV)

the predetermined charge completion level of 0.01C (0.1A). The charging time taken using this method is 5400s (1.5 hrs).

A boost charging profile (Figure 4c) is simulated where a boost-charging period of 5 minutes is applied to the battery. It is a constant voltage step where a high voltage is applied to the battery as a result of which a high current inrush is witnessed in this step due to very high potential difference. Next, the charging profile is switched to normal CC-CV mode to attain rest of the capacity. In this step, a constant current of 1C (10A) is applied to the battery till it attains V_{max} followed by a constant voltage profile at V_{max} to attain 100% SOC. The total charging time taken in this method is 5000s (~ 1.3 hrs).

For each charging profile, simulation has been run for 500 cycles to evaluate the capacity fade at the end of cycling. The results are presented in Table 1:

Table 1: Charging time and capacity fade results of the various charging methods

Charging method	Charging time (s)	Cell capacity after 500 cycles (Ah)
CC-CV	8000	9.06
m(CC-CV)	5400	8.63
Boost	5000	8.42
Proposed SOC based	2500	8.96

The results mentioned in Table 1 clearly indicates the superior performance of the SOC based charging method as the capacity fade is minimal and charging time is greatly reduced compared to other fast charging methods. CC-CV charging shows very little capacity fade but at the cost of very large charging time.

The proposed SOC based charging profile has also been simulated in the lab (Figure 5) using the Bitrode cell tester. A 10Ah (1C rate) LiMn_2O_4 prismatic battery cell from Navitasys, LLC with a nominal voltage of 3.72V has been used for experiments.

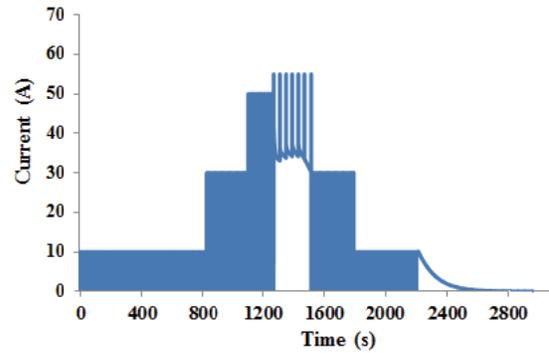


Figure 5. Experimental charging profile performed on a 10Ah LiMn_2O_4 cell

The experimental and simulation results for the charging profile shows very good correlation with each other (Figure 6).

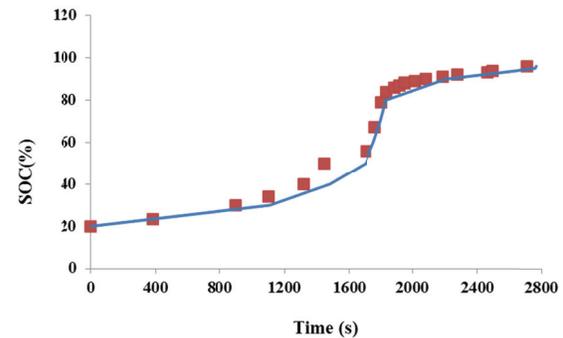


Figure 6. Experimental vs. Simulation result of the SOC based charging profile. Solid line corresponds to simulation data and dotted line represents experimental data

7. Conclusions

The conventional CC-CV charging provides the most optimal charging performance with minimum battery degradation but at the cost of large charging time. The various fast charging methods available in the literature are able to reduce the charging time but are accompanied by large capacity fade as well as safety issues due to overheating as a result of charging at very high currents. The SOC based charging method proposed in this work not only reduces the charging time drastically but also provides optimum battery performance and better thermal management due to controlled charging. This method has been evaluated both by

experimentation and simulation using COMSOL Multiphysics 4.3b.

8. References

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