

Multidimensional Li-Ion Battery Modelization Coupling Thermal and Electrochemical Aspects

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Abstract - Lithium-ion batteries are the preferred choice because of their many advantages, such as higher specific energy and energy densities compared with other secondary battery chemistries for different applications. Substantial diagnostic and modelling efforts are needed to fully understand the thermal and electrical characteristics of lithium-ion cells under various operating conditions. We have developed a multidimensional model that integrates both the thermal and electrochemical aspects of lithium-ion batteries. This model has been designed to perform numerical simulations to analyse heat generation as well as thermal properties such as thermal conductivity and specific heat capacity and their relationship with: battery capacity, charge/discharge rate, ambient conditions and battery geometry. Based on the review presented, a multidimensional Li-Ion battery model coupling thermal and electrochemical aspects is developed and presented. The results of our simulations, carried out using COMSOL, show that our model is capable of accurately predicting the internal temperature distribution of a lithium-ion cell, as well as the evolution of its mean temperature during operation, accordance to the experimental data.

Keywords - lithium-ion (Li-Ion), overheating effect, thermal characterization, Simulation, COMSOL Multiphysics.

I. INTRODUCTION

The process of electrical energy storage has undergone exponential evaluation throughout the last ten years. To run a variety of gadgets (such as cellphones, solar systems, computers, satellites, aeronautical gadgets, robotics, etc.), we require a supply of electrical energy. Reduced emissions of greenhouse gases (GHG) and thus lower levels of global pollution are made possible through energy storage of electricity.

Electrical energy storage is used in these applications to assure operation continuity; the stored energy is then released in accordance with the mission's established requirements [1]. As a result, the production of batteries and the improvement of its systems were always in the news. Recently, materials with better mass-to-energy and volume-to-energy ratios have been used in batteries. Due to its greater energy

density, adaptable form size, and lightweight construction when compared to competing battery technologies, lithium-ion batteries are the most popular energy storage devices.

Due to the complexity of the interactions, it is widely acknowledged that a microscopic description of the battery is unattainable. Early in the 1990s, Newman's group released the first mathematical models detailing the physical characteristics of lithium-ion battery cells [2]. Two macro-homogeneous phases that coexist at each location in the cell are superimposed to depict the porous electrodes. Solid or liquid phases make up these states. Additionally, the solid phase is depicted by spheres that are associated with active material particles (Fig. 1) [3, 4]. Numerous research have revealed that the two primary components of Li-ion batteries' internal heat generation are irreversible heat generation and reversible heat generation.

The focus of this study is on the thermal behavior analysis of lithium-ion 18 650 battery systems under various discharge situations. To simulate operational scenarios typical of these batteries, a number of discharge rates have been established. The surface temperature and tension variations are among the factors that were examined. The main goals of this research are to provide a thorough understanding of thermal risks related to lithium-ion battery operation and to significantly improve the notion of battery security.

II. MODEL DESCRIPTION

There are numerous modeling strategies described in the literature, ranging from straightforward electrical equivalent circuit models to intricate pseudo-two-dimensional electrochemical and thermal models [5]. The main benefits of these physics-based techniques come from the ability to express macro-variables (voltage cell) as a function of microscopic parameters and physical characteristics of the system's components (figure 1). Physical laws including Ohm's Law, Butler-Volmer's Law, and Fick's Diffusion Law are used in electrochemical battery models to depict the internal chemical reactions that take place inside the battery during charging and discharge [6]. Doyle and Newman introduced the original electrochemical battery model.

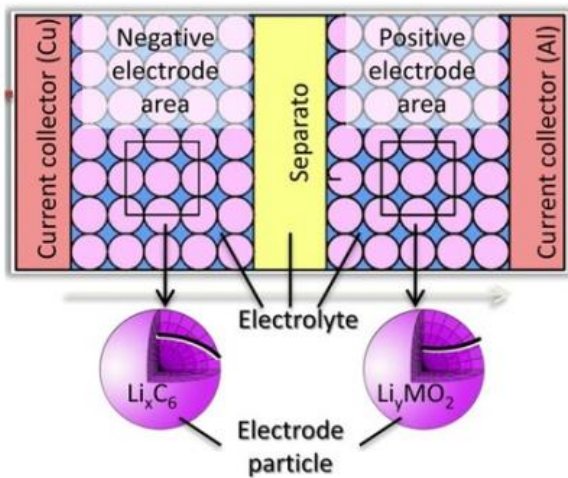


Fig. 1. Schematic macroscopic cell model with coupling solid diffusion model microscopic [7].

III. DESIGN METHODOLOGY AND MODELING UNDER COMSOL

Several critical characteristics of finite element method (FEM) modeling of microsystems must be considered in our research:

- First, demonstrate the benefits of this strategy (complex geometries, non-linearities, multiphysics couplings, and so on).
- The importance of developing multiphysics models to describe the behavior of microsystems, which are systems that integrate many physics (chemical, electrical, thermal, etc.).
- The significance of confirming models generated from the literature or with experimental measurements as much as possible.

IV. MATHEMATICAL MODEL

A) Basic electrochemical model

The model assumes that the active material particles are single-sized spherical particles, and that lithium Li intercalation and deintercalation occur on the surface of the active materials[8]. During the charge-discharge cycle, lithium ions and electrons flow in opposite directions, observing charge conservation; that is, the quantity of lithium ions must equal the electronic charge transfer [9].

- Conservation of Charge

The charge conservation equation in the solid phase is as follows, which is also a form of Ohm's law :

$$\sigma_{s,eff} \left(\frac{\partial^2 \phi_s}{\partial x^2} \right) = a_s F j \quad (1)$$

Where $\sigma_{s,eff}$ is the effective electronic conductivity of the solid phase, ϕ_s is the solid phase potential, F is the Faraday constant and j is the local flux of ions between the pores and the walls at the electrode/electrolyte interface. a_s is the specific surface area of the particles in the solid phase and r_p is related to the particle size radius and the volume fraction of the active particles (ϵ_s):

$$a_s = 3\epsilon_s / r_p \quad (2)$$

The charge conservation equation in the electrolyte phase is shown in the following equation :

$$-K_{l, eff} \frac{\partial \varphi_l}{\partial x} + \frac{2K_{l, eff} RT}{F} \left(1 + \frac{\partial \ln f}{\partial \ln c_l}\right) (1-t_+) \frac{\partial \ln c_l}{\partial x} = i_l \quad (3)$$

$K_{l, eff}$ is the effective ionic conductivity of the electrolytic phase, R is the universal gas constant, F is the Faraday constant, T (K) is the absolute temperature, f is the molecular activity coefficient of the electrolyte, C_l is the concentration in the electrolyte, and t_+ is the Li-ion transfer number.

- Conservation of matter

According to Fick's second law, the diffusion and migration of lithium ions in the solid-phase active materials of the positive and negative electrodes are described by the formula (4), the gradient of the concentration change at the center of the condition formula at the limits (5) is zero, and the reactive ion current is expressed by equation (6).

$$\frac{\partial c_s}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} (D_s r^2 \frac{\partial c_s}{\partial r}) \quad (4)$$

Boundary condition :

$$\frac{\partial c_s}{\partial t} \Big|_{r=0} = 0 \quad (5)$$

$$-D_s \frac{\partial c_s}{\partial t} \Big|_{r=r_p} = \frac{j}{a_s F} \quad (6)$$

Where C_s is solid phase lithium ion concentration, D_s is solid phase diffusion coefficient, r_p is particle radius, t is Li-ion intercalation or deintercalation reaction time, a_s particle surface area of active material, j is the intercalation and deintercalation reaction current, F is Faraday's constant.

The concentrated solution theory is used to model the transport process in the electrolytic phase which depends on the diffusion of lithium ions in the electrolyte C_l , the porosity of the electrode ε_l and the current density of reaction j . In the electrolyte phase, the transport of lithium ions is described as follows:

$$\varepsilon_l \frac{\partial c_l}{\partial t} = D_{l, eff} \frac{\partial^2 c_l}{\partial x^2} + (1-t_+) \frac{j}{F} \quad (7)$$

Charge transfer occurs at the surface of active matter particles in contact with the electrolyte, and the process is described by the Butler-Volmer equation :

$$J = i_0 \left[\exp\left(\frac{\alpha_a F}{RT} \eta\right) - \exp\left(-\frac{\alpha_c F}{RT} \eta\right) \right] \quad (8)$$

The exchange current density i_0 relates to the lithium-ion concentration in the electrode and electrolyte phases, which is defined using the following expression:

$$i_0 = F k C_e^{\alpha_a} (C_{s, max} - C_{s, surf})^{\alpha_a} C_{s, surf}^{\alpha_c} \quad (9)$$

Here, k represents the temperature-dependent reaction rate (subscripts a and c represent anode and cathode, respectively).

The battery overvoltage η is defined as the difference between the actual battery potential and the thermodynamic equilibrium potential U_i as shown below:

$$\eta = \varphi_s - \varphi_l - E_{eq} \quad (10)$$

B) Thermal model

The thermal model is developed to predict the temperature distribution inside the battery using the energy balance equation [10]. Eq (11) is the most commonly used equation for characterizing heat generation in a battery cell during an electrochemical process (charging or discharging).

$$\rho C_p \frac{\partial T}{\partial t} = \nabla(k \nabla T) + Q \quad (11)$$

Where ρ is the density, C_p is the heat capacity at constant pressure, T is the temperature, k is the thermal conductivity and Q is the term that groups together all the sources of heat production.

$$Q = I (E_{oc} - V) - IT \frac{dE_{oc}}{dT} \quad (12)$$

Where Q is the rate of heat generation, I is the applied current ($I > 0$ for discharging and $I < 0$ for charging), E_{oc} is the cell voltage at equilibrium or open circuit potential, V is the cell voltage, T is the temperature and dE_{oc}/dT is the temperature entropy coefficient.

The first term, $I (E_{oc} - V)$ is The irreversible heat generated due to physical process limitations, such as ohmic losses, activation

above potential, and mass transport limitations. While the second term, $IT [dE_{oc}/dT]$ is the reversible heat generated (or absorbed) due to the increased order/disorder in the crystal structure (of both electrodes) due to the insertion/extraction of lithium.

Equations (13) and (14) describe the expressions for heat transfer by convection and radiation respectively, between the environment and the battery surface :

$$(k\nabla T) = h (T_{amb} - T) \quad (13)$$

$$(k\nabla T) = \varepsilon\sigma (T_{amb}^4 - T^4) \quad (14)$$

Where h is the effective heat transfer coefficient and T_{amb} indicates the environment temperature, The heat exchange between the battery and its surroundings is primarily accomplished through three modes of heat transfer: radiation, conduction, and convection. Because the heat delivered via radiation is minimal in comparison to other forms, it can be safely ignored. The heat exchange between the battery and its surroundings is primarily accomplished through three modes of heat transfer: radiation, conduction, and convection. Because the heat delivered via radiation is minimal in comparison to other forms, it can be safely ignored.

V. NUMERICAL SIMULATION PROCESS

In this study, COMSOL Multiphysics 5.4 commercial finite element method (FEM) software was used to solve the coupled electrochemical-thermal model of a commercial cylindrical 18650 format lithium-ion battery with a nominal capacity of 2.6 Ah and LiMn2O4 cathode and carbon anode. The battery cell chemistry was modeled using a one-dimensional cell model, and the temperature in the cell was modeled using a two-dimensional model.

The thermal energy conservation equation uses the rate of heat generation resulting from electrochemical reactions and joule heating to compute the temperature field on a local scale. This temperature data is then given back to the electrochemical system to update it (Fig. 2).

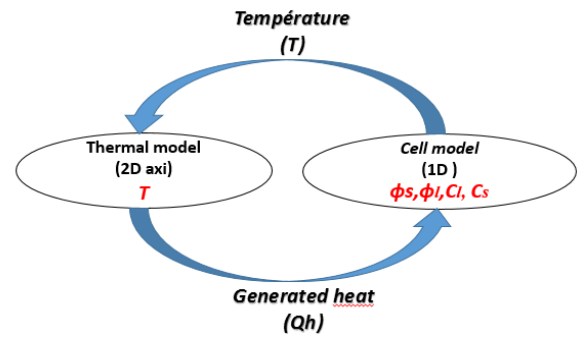


Fig. 2. Coupling between the cell and the thermal model using the average values for the temperature and the heat generated.

Different charge rates (C-rates), such as 0.5C, 1C, 2C, and 4C, were used to charge and discharge the cell. The variation in cell temperature was observed, as well as the variation in thermal contact resistance values between regions of active battery material.

VI. RESULTS AND DISCUSSIONS

The commercial finite element solver COMSOL Multiphysics 5.4 is used to simultaneously solve the electrochemical and thermal model equations. The terminal boundary received an application of I_{app} predetermined battery current. Battery operating voltage, species and concentration distribution, electron and ion current density distribution, and heat generation are examples of model outputs. The created model allows for the retrieval of the cell's potential at any given moment.

A) Evolutions of cell potential

Figure 3 illustrates the fluctuation in discharge capacity at various C-rate rates, including 0.5C, 1C, 2C, and 4C, at a 25°C ambient temperature. Due to the increase in internal resistance at the end of the discharge process (i.e. the resistance of solution, the contact resistance of the electrode to the current collector, the kinetic resistance, and the diffusion resistance), the discharge capacity consequently decreases significantly. It should be noted that the battery voltage drops faster as the C-rate increases. Additionally, the battery discharge capacity is 50% of the theoretical capacity when we examine at a rate of 4C.

The positive electrode was loaded with Li during the discharge, as evidenced by the significant increase at the end of the discharge, and the opposite is true at the conclusion of the charge for the negative electrode.

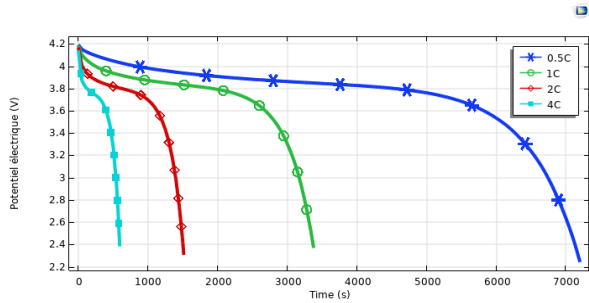


Fig. 3. Voltage delivered by the cell for different solicitation currents

Figure 4 illustrates the battery capacity decreases as the temperature drops, in part because the platform life and battery voltage are shortened; this can be seen at 10°C and decrease to temperature 0°C and, as the battery temperature drops, the lithium ion diffusion activity of the cathode gradually reduced. Second, the internal resistance of the battery increases over time, increasing both the internal pressure and the time required to reach the cutoff voltage.

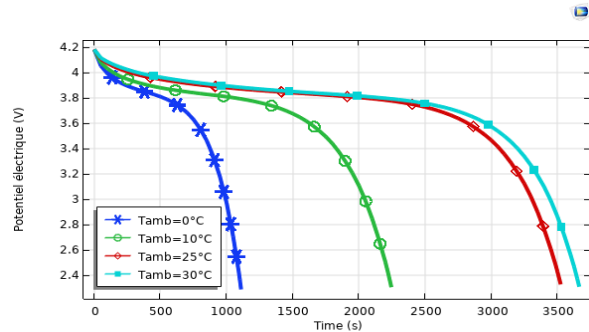


Fig. 4. Discharge with 1C rate starting from various temperatures.

B) Effect of C-Rate on Heat Generation of Battery

In the following figure (Fig. 5), which shows the cell being asked by constant current regimes at 0.5C, 1C, 2C, and 4C in discharge, it is discovered that the regime has a significant impact on the accumulator's thermal behavior.

The battery will generate more heat as the current rate increases, and this heating will be significant. However, for high C rates, this behavior is not linear. As the discharge process came to a close, the average cell temperature increased significantly. This was most likely brought on by a significant polarization that occurred during the discharge procedure.

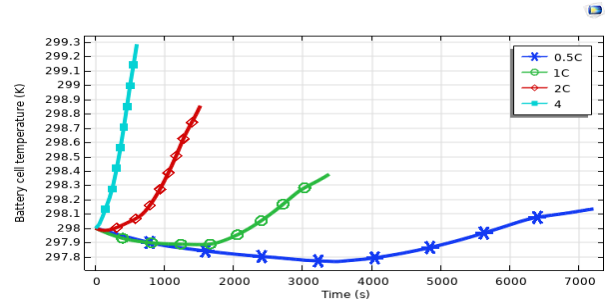


Fig. 5. Influence of temperature for different solicitation currents.

Temperature has a positive correlation with discharge current, as expected. As a result, it is clear from Figure 6 that increasing the discharge rate causes an increase in temperature, which is due to an increase in heat production. The maximum temperature appears at the active material level. Indeed, the electrochemical reaction raises the internal heat generation rate of the battery while decreasing external convection.

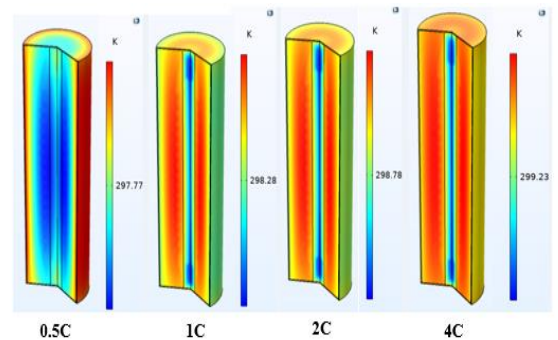


Fig. 6. Internal temperature distribution of batteries with different rates (at Time=3000s).

VII. CONCLUSION

A lithium cell electrothermal behavior model was discussed for a Li-ion battery to understand its performance characteristics at different charge/discharge rates. For higher rates of C passing through the cell, this results in more

movement of Li⁺ ions from an electrode to electrode, which generates more heat in the cell. Under load, two main heat sources are generally used to model the thermal behavior of a Li-ion battery. The chemical character of Li-ion elements is strongly impacted by the electrical stress applied to them on the one hand, but also by the temperature which causes their Physico-chemical properties to vary on the other hand. Their thermal behavior can thus be modified.

The prediction of the thermal behavior of a Li-ion accumulator can only be made from the coupling between a fairly complex electrochemical model and a thermal model. The intrinsic relationship between these two types of phenomenon does not allow the realization of a purely predictive thermal model, without the need to experimentally characterize the accumulator. It is apparent from this simulation work that the presence of thermal contact resistances between the cell layers is one of the reasons for the generation of heat inside the cell.

Currently, the focus of our research is on investigating various cooling techniques, such as forced air-cooling and the use of phase-change materials (PCM). Passive thermal management systems are employed to maintain battery temperature uniformly within the phase change temperature range, all without the need for additional energy consumption.

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