Journal of Power Sources 290 (2015) 87–101

Contents lists available at ScienceDirect

Journal of Power Sources

journal homepage: www.elsevier.com/locate/jpowsour

A reduced order electrochemical thermal model for lithium ion cells

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HIGHLIGHTS

- Reduced order model for coupled electrochemical thermal response
- Thermal balance with local heat generation used for model order reduction.
- Model validated with experimental data at low, high and room temperatures.
- Model validation with HPPC and UDDS data.

ARTICLE INFO

Article history:
Received 21 February 2015
Received in revised form
27 April 2015
Accepted 30 April 2015
Available online 15 May 2015

Keywords:
Lithium ion cells
Thermal model
Nickel cobalt aluminium oxide
Reduced order model

ABSTRACT

A reduced order model (ROM) is proposed for accurate prediction of electrochemical and thermal response of lithium ion cells. The order reduction is carried on the coupled partial differential equations (PDE) of the electrochemical thermal model by consistent volume averaging of local heat generation and spatial temperature variation. The model is validated with experimental data for temperatures ranging from 253 K–333 K. It is seen that modification of ROM to account for low electronic conductivity results in accurate voltage estimation of cells with lithium nickel cobalt aluminium oxide (LNCO) cathodes. A detailed parametric sensitivity to operating conditions is provided. The utility of ROM for on-board state estimation is demonstrated by applying to realistic drive cycle protocols such as the Hybrid Pulse Power Characterization (HPPC) and the Urban Dynamometer Driving Schedule (UDDS) data. The electrochemical structure of ROM enables identification of controlling processes, and analysis of HPPC results reveal that Ohmic drop of cathode is controlling at high rates and the electrolyte potential during rest phase. Based on accurate voltage prediction, computational speed and physical insights, it can be concluded that the proposed ROM is an adequate state estimation and a cell design tool.

1. Introduction

The foray of electric and hybrid electric vehicles into the commercial automobile market is largely due to the lithium ion batteries which provide high energy/power density with stable cycle performance. Although it directly results in a driving range almost comparable to mainstream automobiles, the high energy density of lithium ion batteries also results in increased concerns of thermal safety. Studying the thermal behavior of these cells is therefore, becoming an active area of research.

Nickel oxide based electrodes are quite commonly used in lithium ion cells. Having an accurate real-time state estimation model is increasingly important when this electrode material is used, as its low thermal stability is a serious concern. Chen et al. [1] first studied that doping nickel cobalt oxide with aluminium helps in stabilizing the impedance rise otherwise seen in the material. This paved way for lithium nickel cobalt aluminium oxide (LNCAO) to be used as the positive electrode for high power cells recommended for application in hybrid electric vehicles. The thermal characteristics of LNCAO/carbon (C) cell have been the focus of numerous studies. Kobayashi et al. [2] find that the capacity loss of the LNCAO/C cell is significantly higher upon cycling at high temperature. This is corroborated by Kim et al. [3] by obtaining a differential scanning calorimetry peak at a relatively low temperature

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http://dx.doi.org/10.1016/j.jpowsour.2015.04.179
0378-7753/

of ~208 °C for LNCAO cells. Rad et al. [4] propose a thermal model for LNCAO cells. Although the model is validated with experimental data from 10–40 °C contributions from entropic and overpotential heat are alone considered while the low temperature behavior is not considered.

Each of the electrochemical and transport processes occurring within the cell act as heat sources contributing to the cell temperature. As these processes themselves are affected by factors such as current rates and ambient temperatures, the cell temperature can vary significantly during operation. To avoid scenarios such as thermal runaway, predictive models for the cell temperature are necessary. The electrochemical model combined with a thermal balance is one of the approaches of obtaining a predictive thermal model for lithium ion cells. The advantage of using a physics based model is that system-specific heat sources, such as phase change, can be easily included within the model.

In one of the earliest works on thermal modeling of lithium ion cells, Pals and Newman [5] study the thermal behavior of a lithium polymer cell by combining the electrochemical model equations given by Doyle et al. [6] with the general energy balance proposed by Bernardi et al. [7]. Gu and Wang [8] study the two dimensional thermal behavior of a lithium metal oxide cell and demonstrate its advantages over a decoupled and an isothermal model. In addition to using a similar approach, Srinivasan and Wang [9] include the contribution from reversible heat for modeling the thermal behavior of a lithium manganese oxide cell. This is achieved by including contribution from the entropic heat from electrodes into a lumped thermal equation. Kumaresan et al. [10] argue that with a change in the cell temperature, properties such as the diffusion coefficient, ionic conductivity, non-ideal nature of the electrolyte phase and the diffusion coefficient of the solid phase vary substantially. Therefore, a systematic study of the effect of varying each of the above parameter with temperature is carried out. A significant deviation in the end of discharge characteristics is observed when the system properties are considered with and without the temperature dependence corroborating the importance of including it.

Following the development of the coupled electrochemical thermal model, Ye et al. [11] carry a detailed validation of it with experimental data at various temperatures and charge/discharge rates. The model is used to obtain lithium concentration profiles within the cell which in turn is used to obtain cell design guidelines for better utilization. Cleary et al. [12] use the electrochemical thermal model to obtain local cell potential and temperature information which is input to a 3D model of spiral and prismatic cells. Mao et al. [13] simulate the temperature rise within the lithium ion cell when it is subjected to high current rates making use of the dual foil model [6]. Zhao et al. [14] use the thermal electrochemical model to study the thermal behavior of lithium manganese oxide cells. The study investigates the significance of the contribution of reversible heat to the total heat generated with spatial resolution, that is, along the thickness of the cell. Ye et al. [15] propose a two dimensional electrochemical thermal model suitable for large format lithium ion cells. The cross plane temperature gradients are studied using a modified thermal conductivity which takes into account the thermal contact resistance between the cell layers. It is shown that the external cooling is insufficient to mitigate the high temperatures within the cell interiors and battery designs which can mitigate these cross-plane temperature gradients are suggested. Tourani et al. [16] study the battery cell response to the driving cycles. The findings are used to suggest better guidelines for thermal management of the battery cells in practical use scenarios.

The discharge capacity of the lithium ion cells drops significantly at low temperatures and it becomes increasingly difficult to achieve complete capacity upon charging. Accurate state of charge estimation of the cells in such ambient conditions is highly important and at the same time equally difficult. The atypical behavior of these cells at low temperatures has therefore been the focus of various studies and is attributed to slowing of the electrochemical and transport processes within the cell. Zhang et al. [17] study the low temperature performance of the lithium ion cells using electrochemical impedance spectroscopy. The main contributing factor to the low temperature capacity drop is the increased charge transfer resistance which in turn originates from the slow reaction kinetics at these temperatures. In contrast, Ji et al. [18] attribute the low temperature performance to lithium diffusion within the negative electrode and electrolyte phase. Yi et al. [19] provide a model applicable for low temperature operation of lithium ion cells. Polynomial expressions are obtained via fitting with experimental data for two parameters which modify the current density.

Despite including intricate physical details, extending it to 3 dimensions and low temperatures, the electrochemical thermal model needs to be amenable for onboard implementation. With this view, Cai and White [20] use the proper orthogonal decomposition method to obtain a reduced order finite volume representation of the rigorous electrochemical thermal model. The reduced model computes 70% faster while the results compare well with the complete model solved in COMSOL. Guo et al. [21] combine the lumped thermal model equation [7] with the single particle model. The solution phase polarization is taken into account by including a resistance term. A study and analysis of the heat generation through various sources such as reversible and irreversible is carried out. Prada et al. [22] also make use of the lumped thermal model by combining it with the average model which is a simplified form of the complete electrochemical model. Brown and Landers [23] make use of the single particle model with the lumped thermal balance equation to study the effect of the cooling fluid temperature and flow rate on the cell temperature. The model is further simplified by linearizing the equations and using the Arnoldi algorithm to make it amenable for onboard implementation. Remmlinger et al. [24] use a linear parameter varying model to reduce the order of the electrochemical model.

In contrast to the discussed reduced models which use intricate mathematics or the simplified single particle model, Kumar [25] and Gambhire et al. [26] use the complete electrochemical model. The model order reduction is carried out using volume averaging and the spatial information is obtained by suitable profile approximations. Although this model order reduction involves simple methods and includes the detailed physics of the electrochemical model it is valid only for isothermal operation of cells.

For accurate prediction of cell behavior while undergoing realistic drive cycles, in the present work, a reduced order model (ROM) is proposed to study the electrochemical and thermal response of a lithium ion cell. The model order reduction is carried out on the electrochemical model retaining the physics based nature. In addition, the distributed thermal balance equation [27] is used which takes into account the individual contribution of the various heat sources within the cell unlike the previous reduced order models where the lumped thermal balance equation is used. The temperature and concentration dependence of the transport properties are also considered in a consistent manner in the proposed model. This provides the present ROM with the dual advantages of being amenable for onboard usage while providing physical insights and design guidelines for the lithium ion cells.

The manuscript is structured as follows. Details of experiments conducted on a commercial LNCAO/C cell are described in section (2). Section (3) includes the detailed derivation of the reduced order coupled electrochemical thermal model. Section (4) includes the results obtained from the model beginning with the assessment