International Journal of Heat and Mass Transfer 117 (2018) 261–272

Contents lists available at ScienceDirect

International Journal of Heat and Mass Transfer

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

# Numerical modeling and analysis of the thermal behavior of NCM lithium-ion batteries subjected to very high C-rate discharge/charge operations

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ARTICLE INFO

*Article history:*
Received 5 May 2017
Received in revised form 15 September 2017
Accepted 6 October 2017

*Keywords:*
Lithium-ion battery
Thermal behavior
Very high C-rate operation
Thermal runaway
Battery thermal management system

ABSTRACT

Lithium-ion batteries are easily overheated during discharge/charge operations with large current output/input. Traditional battery tests are difficult to pinpoint the internal thermal mechanism for an overheated battery. In this study, it is proposed a model to investigate the thermal behavior of the charge and discharge processes of lithium-ion battery with very high C-rate. The model combines an electrochemical-thermal (ECT) coupled module and a thermal abuse module. The whole successive process of the cell operation including charge/discharge, battery material exothermic reactions, and even thermal runaway within a cell, is fully described, by a single model. Predictions of individual $LiNi_xCo_yMn_zO_2$ (NCM) lithium-ion cell high C-rate (up to 8C) discharge/charge processes compare well with experimental data. A detailed analysis is conducted to evaluate the influence of external heat release condition and charge/discharge C-rate on the thermal behavior of batteries during and after very high C-rate (>8C) charge/discharge operations. Results indicate: (1) the very large output/input current leads to the early-coming of cut-off voltage, terminating the discharge/charge operation; (2) compared with the very high C-rate charge operation, the discharge operation of the same C-rate is easier to cause battery overheat, leading to the occurrence of battery thermal runaway; (3) the high C-rate charge operation with cut-off voltage control fault is very dangerous as it can cause very fast heat generation and eventually possible thermal runaway; (4) favorable heat release condition or effective and active thermal control may be the key to the thermal control and restraining thermal runaway of lithium-ion batteries.



## 1. Introduction

Lithium-ion batteries (LIBs) are common in a variety of energy storage applications. Batteries with $LiNi_xCo_yMn_zO_2$ (NCM) or $LiNi_xCo_yAl_zO_2$ (NCA) cathode are widely used in electrical vehicles (EVs), such as BMW i3, Chevrolet Volt, Nissan Leaf, and Tesla Model S and Model 3. The frequent incidents including combustion and explosion of LIBs indicate that the safety issue persists to be one major challenge that prevents the large-scale commercialization of EVs. Overheat of battery in the discharge/charge process is one of the main causes for these incidents [1].

During high or very high C-rate charge/discharge processes, LIBs are more vulnerable to overheat because a large amount heat is generated in the cell at a very high rate; the overheat of battery may feed the subsequent abusive reactions of battery materials causing further overheat of the battery, which, in turn, may result in thermal runaway or even incidents like combustion and explosion [2]. The EVs applications of LIB require the battery to tolerate relatively high C-rate discharge/charge operations. Therefore, considerable research on the thermal behavior of LIBs during high or very high C-rate discharge/charge operations is a must.

Numerous publications dealing with the thermal behavior of LIBs during charge/discharge processes have appeared in the open literature. Quadir et al. [3] measured the overpotential and entropic heat generation and calculated the heat generation in a Li-ion cell during 1C-4C discharge processes. Drake et al. [4] measured the cell temperature and surface heat flux to determine the heat generation rate in batteries with high discharge rates, up to 9.6C. The detailed temperature distribution across the surface of a large format 20 Ah pouch cell was investigated over a wide ranges of

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https://doi.org/10.1016/j.ijheatmasstransfer.2017.10.024
0017-9310/

**Nomenclature**

*List of symbols*

| | |
|---|---|
| $A$ | side surface area of the electrode plate ($m^2$) |
| $A_{sei}$ | SEI decomposition frequency factor ($s^{-1}$) |
| $A_{ne}$ | negative-solvent frequency factor ($s^{-1}$) |
| $A_{pe}$ | positive-solvent frequency factor ($s^{-1}$) |
| $A_e$ | electrolyte decomposition frequency factor ($s^{-1}$) |
| $A_s$ | side surface area of the electrode plate ($m^2$) |
| $a_s$ | specific surface area ($m^{-1}$) |
| $c$ | $Li^+$ concentration (mol $m^{-3}$) |
| $c_{ele}$ | dimensionless concentration of electrolyte |
| $c_{neg}$ | dimensionless amount of lithium within the carbon |
| $c_p$ | specific heat capacity (J $kg^{-1}$ $K^{-1}$) |
| $c_{sei}$ | dimensionless amount of lithium containing metastable species in the SEI |
| $D$ | diffusion coefficient ($m^2$ $s^{-1}$) |
| $E_{a,sei}$ | SEI decomposition activation energy (J $mol^{-1}$) |
| $E_{a,ne}$ | negative-solvent activation energy (J $mol^{-1}$) |
| $E_{a,pe}$ | positive-solvent activation energy (J $mol^{-1}$) |
| $E_{a,e}$ | electrolyte decomposition activation energy (J $mol^{-1}$) |
| $F$ | Faraday's constant (C $mol^{-1}$) |
| $h$ | equivalent heat transfer coefficient ($Wm^{-2}$ $K^{-1}$) |
| $H_{sei}$ | SEI-decomposition heat (J $kg^{-1}$) |
| $H_{ne}$ | negative-solvent reaction heat (J $kg^{-1}$) |
| $H_{pe}$ | positive-solvent reaction heat (J $kg^{-1}$) |
| $H_e$ | electrolyte decomposition heat (J $kg^{-1}$) |
| $I$ | current load (A) |
| $i_o$ | exchange current density (A $m^{-2}$) |
| $j^{Li}$ | transfer current density (A $m^{-3}$) |
| $L$ | through-plane thickness of the battery |
| $L_e$ | through-plane thickness of the electrode (m) |
| $m_{sei}$ | reaction order for $c_{sei}$ |
| $m_{ne,n}$ | reaction order for $c_{neg}$ |
| $m_{pe,p1}$ | reaction order for $a$ |
| $m_{pe,p2}$ | reaction order for (1-$a$) |
| $m_e$ | reaction order for $c_e$ |
| $p$ | Bruggeman factor |
| $q$ | volumetric heat generation rate |
| $q_c$ | contact resistance heat (W $m^{-3}$) |
| $q_{ele}$ | heat from electrolyte decomposition reaction (W $m^{-3}$) |
| $q_i$ | ionic ohmic heat (W $m^{-3}$) |
| $q_{ir}$ | heat from irreversible electrochemical reaction (W $m^{-3}$) |
| $q_{ne}$ | heat from reaction between negative active material and electrolyte (W $m^{-3}$) |
| $q_o$ | ohmic heat (W $m^{-3}$) |
| $q_{pe}$ | heat from reaction between positive active material and electrolyte (W $m^{-3}$) |
| $q_{re}$ | reversible entropic heat (W $m^{-3}$) |
| $q_{sei}$ | heat from the SEI decomposition reaction (W $m^{-3}$) |
| $Q_1$ | total heat generation rate form ECT model (W $m^{-3}$) |
| $Q_2$ | total heat generation from exothermic reactions of battery materials (W $m^{-3}$) |
| $r$ | radius of solid active particles (m) |
| $R_c$ | the total lumped contact resistance ($\Omega$ $m^2$) |
| $R_{sei}$ | SEI-decomposition reaction rate ($s^{-1}$) |
| $R_{ne}$ | negative-solvent reaction rate ($s^{-1}$) |
| $R_{pe}$ | positive-solvent reaction rate ($s^{-1}$) |
| $R_e$ | electrolyte decomposition reaction rate ($s^{-1}$) |
| $T$ | temperature (K) |
| $T_{amb}$ | ambient temperature |
| $T_{ref}$ | reference temperature (K) |
| $T_{sur}$ | temperature at the cell surface (K) |
| $t$ | time (s) |
| $t_e$ | characteristic time |
| $t_s$ | characteristic time |
| $t_c$ | characteristic time |
| $t_+^0$ | transference number of Li dissolved in the electrolyte |
| $t_{sei}$ | dimensionless measure of SEI layer thickness that reflects the amount of lithium in the SEI |
| $U$ | open-circuit potential (V) |
| $V$ | cell voltage (V) |
| $v$ | volume of numerical element interfacing separator and electrode ($m^{-3}$) |
| $W_c$ | volume-specific carbon content before abusive reactions (kg $m^{-3}$) |
| $W_p$ | volume-specific active material content in cathode before abusive reactions (kg $m^{-3}$) |
| $W_e$ | volume-specific electrolyte content before abusive reactions (kg $m^{-3}$) |

*Greek symbols*

| | |
|---|---|
| $\alpha$ | degree of conversion |
| $\alpha_a$ | anodic transfer coefficient |
| $\alpha_c$ | cathodic transfer coefficient |
| $\varepsilon$ | porosity |
| $\eta$ | surface overpotential (V) |
| $\kappa$ | ionic conductivity (S $m^{-1}$) |
| $\lambda$ | thermal conductivity (W $m^{-1}$ K) |
| $\rho$ | density (kg $m^{-3}$) |
| $\sigma$ | electronic conductivity (S $m^{-1}$) |
| $\varphi$ | electric potential (V) |
| $\delta$ | thickness of copper collector (m) |

*Subscripts/superscripts*

| | |
|---|---|
| 0 | initial value |
| e | electrolyte phase |
| eff | effective value |
| neg | negative electrode |
| pos | positive electrode |
| s | solid phase |
| sep | separator |

ambient temperature and discharge/charge C-rate (0.5C-10C) [5]. The core temperature of a Li-ion cell was indirectly determined by integrating spatio-temporally the measured temperature field on the outside surface of the battery during 2C-10C discharge processes [6]. However, the thermal behavior of Li-ion batteries during higher C-rate (particularly >10C) operations, which probably causes battery destruction or even thermal runaway, is rarely studied.

Numerical models are also widely used to study the involved thermal behavior characteristics of batteries during charge/discharge operations of various C-rates. The established or employed models can be classified as: electrochemical-thermal (ECT) coupling model [2,7–14], electro-thermal (ET) coupling model [15,16], equivalent circuit model (ECM) [10], and partially coupled model [17,18]. The simulated discharge/charge operations were mostly below 10C except the work by Cai and White [11], in which they modeled the 0.1C-20C discharge processes and found that the maximum cell temperature rise (70 °C) was harvested at the interruption time of 10C discharge operation when the cut-off voltage (3.0 V) was reached.

Super high C-rate (up to 50C or even 397C) discharge processes were explored with respect to batteries of special cathode materi-