



Combined experimental and numerical evaluation of the differences between convective and conductive thermal control on the performance of a lithium ion cell



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ABSTRACT

Testing of lithium ion batteries is necessary in order to understand their performance, to parameterise and furthermore validate models to predict their behaviour. Tests of this nature are normally conducted in thermal/climate chambers which use forced air convection to distribute heat. However, as they control air temperature, and cannot easily adapt to the changing rate of heat generated within a cell, it is very difficult to maintain constant cell temperatures. This paper describes a novel conductive thermal management system which maintains cell temperature reliably whilst also minimising thermal gradients. We show the thermal gradient effect towards cell performance is pronounced below operating temperature of 25 °C at 2-C discharge under forced air convection. The predicted internal cell temperature can be up to 4 °C hotter than the surface temperature at 5 °C ambient condition and eventually causes layers to be discharge at different current rates. The new conductive method reduces external temperature deviations of the cell to within 1.5 °C, providing much more reliable data for parameterising a thermally discretised model. This method demonstrates the errors in estimating physiochemical parameters; notably diffusion coefficients, can be up to four times smaller as compared to parameterisation based on convective test data.

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1. Introduction

Thermal or climate chambers are ubiquitous for battery testing to maintain controllable thermal boundary conditions. They are used for studies to investigate degradation and ageing, overall cell performances and its reliability [1–3]. In doing so, they are often used to create data to validate and parameterise models, with the latter often involving fitting the model to the experimental data in order to extract parameters that cannot be measured directly. However, in most cases the model has been assumed to be isothermal and hence any deviation from this assumption in the test data will cause errors in the parameter fitting and validation of the model [4]. To apply isothermal conditions in a battery model is easy, however to achieve the isothermal conditions during testing is relatively difficult. It is the magnitude of the error that this

assumption induces that is really of interest. This assumption can be valid for instance during electrochemical impedance spectroscopy (EIS) measurements taken with low currents at different open circuit potentials. During EIS measurement, it is highly unlikely for a cell to generate significant amount of heat and therefore this assumption will not introduce significant errors. However, during charging and discharging at C rates approaching 1 or more, this condition is surprisingly difficult to achieve. At high currents operation, heat generated will be significant and the cell will be at different temperature from the target temperature. The errors which come from this assumption are rarely discussed or taken into consideration and instead models are fitted to the data on the assumption that the cell is at the temperature of the thermal chamber [5,6]. From a battery modelling perspective, convection coefficient is required typically in a non-isothermal condition. This parameter will change the surface temperature of the battery particularly for active/passive thermal control [7,8]. Therefore by having parameters that are fitted at its true temperature, will eventually give better prediction regardless of the nature of cooling/heating mechanism.

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Nomenclature	
A	effective electrode area [cm^2]
A_s	battery surface area [cm^2]
a_s	specific interfacial surface area [$\text{cm}^2 \text{cm}^{-3}$]
C_e	volume averaged concentration of lithium in electrolyte [mol cm^{-3}]
C_s	volume averaged concentration of lithium in solid [mol cm^{-3}]
D_e^{eff}	electrolyte phase diffusion coefficient [$\text{cm}^2 \text{s}^{-1}$]
D_e	electrolyte phase diffusion coefficient [$\text{cm}^2 \text{s}^{-1}$]
D_s	solid phase diffusion coefficient [$\text{cm}^2 \text{s}^{-1}$]
E_{act}	activation energy for various physiochemical properties [J mol^{-1}]
F	Faraday's constant [C mol^{-1}]
h	convective heat transfer coefficient [$\text{W cm}^{-2} \text{K}^{-1}$]
I	applied current [A]
j_{Li}	reaction current density [A cm^{-3}]
k_y	effective thermal conductivity in through plane direction [$\text{W cm}^{-1} \text{K}^{-1}$]
k_{ct}	charge transfer coefficient [$\text{cm}^{2.5} \text{mol}^{-0.5} \text{s}^{-1}$]
L_n	length of negative electrode [cm]
L_p	length of positive electrode [cm]
l_{cell}	distance between cell unit [cm]
q_c	heat from internal contact resistance [W]
q_j	ionic ohmic heat [W]
q_r	heat from reaction current density [W]
R	gas constant [$\text{J mol}^{-1} \text{K}^{-1}$]
R_f	current collector contact resistance [Ωcm^2]
R_s	radius of active material particle [cm]
t	time
t^{0+}	transference number
T_1	cell surface temperature [K]
T_2	internal cell temperature [K]
T_∞	controlled ambient temperature [K]
U^+	cathode potential [V]
U^-	anode potential [V]
V	cell terminal voltage [V]
v^{vol}	cell volume [cm^{-3}]
x_{stoi}	anode stoichiometric
y_{stoi}	cathode stoichiometric
<i>Greek letters</i>	
δ	electrode thickness [cm]
ϕ_e	electrolyte phase potential [V]
ϕ_s	solid phase potential [V]
ρ	bruggeman coefficient
Θ'	non-dimensional capacity
ε_e	electrolyte volume fraction
ε_s	solid active material volume fraction
ρC_p	volumetric heat capacity [$\text{J cm}^{-3} \text{K}^{-1}$]
<i>Abbreviations</i>	
BMS	battery management system
ECM	equivalent circuit model
EIS	electrochemical impedance spectroscopy
EV	electric vehicle
GITT	galvanostatic intermittent titration technique
HEV	hybrid electric vehicle
OCV	open circuit voltage
SOC	state of charge
SOH	state of health

A lithium ion cell is made from alternating layers of positive electrode, separator, negative electrode and current collectors with each of them having appreciable differences in thermal conductivities and non-trivial thermal contact resistances between them. Since all the materials are layered together with most materials having low thermal conductivity, the effective thermal conductivity in the through-plane direction, depending on cell geometry can be greater than an order of magnitude less than the in-plane direction [9,10]. Cooling of a cell in a thermal chamber can occur via two principle surfaces, the cell surface with a large surface area and short distances but poor through-plane thermal conductivity (i.e. the thickness of a pouch cell is significantly shorter than the length or width), or the cell tabs with excellent in-plane thermal conductivity via the current collectors but long distances and a small surface area (unless high surface area heat sinks are attached). Which one dominates is difficult to be quantified when designing an experiment in a thermal chamber. Forced convection is typically employed for cooling via cell surfaces. This cooling method is widely used for thermal management in a battery pack. The thermal management of battery pack often requires the cooling mechanism to be able to keep the batteries within the operating temperature and minimise the temperature non-uniformity between cells. However, at this level, it is inherently difficult to keep each of the battery to be thermally identical by just relying on the forced convection. The introduction of phase change material which wraps the batteries seem to be a viable solution to reduce temperature increase and keep each of the cells to be relatively identical in terms of its surface temperature [8,11,12]. Nevertheless, the phase change materials is not effective in removing heat from

internal region of the battery primarily due to low rate of heat removal in the through-plane direction.

Chen et al. [13] produced a three dimensional cell thermal model and showed that regions which have high thermal conductivity have more uniform temperature profiles and Jung and Kang [14] also highlighted the effect of high thermal conductivity on surface temperature uniformity. However the poor thermal conductivity in the through-plane direction cannot simply be ignored, for instance [15–17] presented comprehensive cell models but since they ignored this effect, their models will be inaccurate as they would not be able to predict thermal gradients in the through-plane direction. Feng et al. [18] conducted a nail penetration test to evaluate thermal runaway propagation and showed that the local temperature at the battery and internal surface are significantly distinct. A three-dimensional battery model was also made to facilitate the understanding of the occurrence of non-homogeneous temperature. The occurrence of thermal gradient particularly in the through plane direction is primarily due to the low thermal conductivity in this direction. Confirming the presence and magnitude of these thermal gradients is very important and hence many attempts have been made to measure the internal temperature of a cell. However this is difficult to achieve without changing the behaviour of the cell to be studied as shown by Zhe et al. [16]. Several attempts have been made to measure internal temperatures by installing thermocouples inside a cell and significant temperature deviation from cell surface temperature have been recorded [16,19,20]. Richardson et al. [19] used embedded thermocouples to show that combining EIS measurements with measured surface temperature, the internal temperature can be

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