Lithium ion batteries are increasingly important in large scale applications where thermal management is critical for safety and lifetime. Yet, the effect of different thermal boundary conditions on the performance and lifetime is still not fully understood. In this work, a two-dimensional electro-thermal model is developed to simulate cell performance and internal states under complex thermal boundary conditions. Attention was paid to model, not only the electrode stack but also the non-cell components (e.g. tab weld points) and thermal boundaries, but also the experiments required to parameterize the thermal model, and the reversible heat generation. The model is comprehensively validated and the performance of tab and surface cooling strategies was evaluated across a wide range of operating conditions. Surface cooling was shown to keep the cell at a lower average temperature, but with a large thermal gradient for high C rates. Tab cooling provided much smaller thermal gradients but higher average temperatures caused by lower heat removing ability. The thermal resistance between the current collectors and tabs was found to be the most significant heat transfer bottleneck and efforts to improve this could have significant positive impacts on the performance of Li-ion batteries considering the other advantages of tab cooling.

Modeling the Effects of Thermal Gradients Induced by Tab and Surface Cooling on Lithium Ion Cell Performance

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Concerns over internal combustion engine (ICE) emissions have driven a dramatic increase in the uptake of electric vehicles (EVs) in recent years. Despite the EV stock surpassing 2 million vehicles in 2016, it still only accounts for a relatively small fraction of the total vehicle stock.1 To increase EV uptake, EV batteries, which are primarily made up of lithium-ion (Li-ion) cells, must last longer and cost less.

Temperature is one of the key limiting factors for battery pack performance and lifetime, where large temperature deviation from ambient (approximately 20°C) could lead to de-rating, accelerated degradation and in extreme cases, thermal runaway.2–4 Thermal management systems (TMS) are employed in majority of vehicles to counter these challenges.5 Cells can be cooled at different surfaces, namely the electrode surfaces or the electrical connection tabs, or a combination of both. Different liquids or air can be used as the cooling medium, and both can be applied directly or indirectly,10,14. The TMS is employed to keep the temperature of the cells in a defined range where the degradation is minimized and the performance is maximized. However, due to heat transfer limitations, thermal gradients will develop between cells in a battery pack as well as within each cell under aggressive usage,15,16 which has been shown to have an adverse effect on Li-ion cell performance. We have previously studied the effect of TMS choice on the Li-ion cell performance and degradation, where it was shown that surface cooling can cause a higher rate of useable capacity loss compared to tab cooling.17 It was hypothesized that thermal gradients between electrode layers within a cell led to current inhomogeneities and consequently faster power fade. The in-situ measurement of internal states such as current, temperature and state of charge (SoC) is difficult to achieve without disrupting the cell.18–20 Therefore, a model-based approach is required to evaluate the current, temperature, and SoC inhomogeneity induced by thermal gradients.

A useful battery model needs to accurately capture the electrical performance of the cell as well as the electro-thermal interactions in order to be able to provide a spatial resolution of temperature distribution inside the cell. There are broadly two approaches in modeling a lithium ion cell, namely the physics-based pseudo 2-dimensional (P2D) model and empirical equivalent-circuit network (ECN) models. The most commonly used physics-based model is based upon those developed by Newman et al., which use porous electrode and concentrated solution theories.21,22 This type of battery model produces a versatile framework to simulate the Li-ion cell performance over a wide range of operating conditions. However, the computational speed is one of the major challenges when scaling this model from a lumped cell model to a multi-dimensional model with multiple electrochemical models.23,24 Guo et al. showed that the computational speed can be significantly shortened by performing model order reduction.25 However, the model still requires over 30 parameters related to the cell geometry, salt concentration and material properties.26,27 The parameterization is complex and requires access to specialized equipment, which are costly and difficult to access for most end users. For these reasons, despite their advantages, P2D models are not used this work.

On the other hand, ECN models simulate the current-voltage behavior of the Li-ion cell by using a network of resistors, capacitors and voltage sources.28–30 The model is widely adopted by the control and system engineering communities due to its simplicity and robustness within the training data. The work pioneered by Newman, Tiedemann, Gu and Kwon (NTGK) et al.31–34 have shown that a simple ECN or polynomial cell model can be scaled up to a two or three-dimensional model that may be desirable for large format cells. These models solve the potential and current distribution within the electrode plane. In addition, the model can predict the State of Charge (SoC) distribution within the electrode plane or through the cell stack. The cell model parameterization only requires standard procedures such as Pulse Discharge (PD) or Electrochemical Impedance Spectroscopy (EIS). Due its simplicity, computational speed and ease of parameterization the distributed ECN model approach is adopted in this work.

There is a wide range of work in the literature focusing on thermal modeling of Li-ion batteries. The simplest and the most common approach is to treat the cell as a homogenous and isotropic material.35 The limitation of this approach is that the local thermal gradient cannot be accurately reflected due to the isotropic assumption. Some higher dimensional models resolves the cell components such as the electrodes and current collectors individually, thereby capturing the anisotropic heat transfer.36,37 However, with only a few exceptions36,38 existing Li-ion models often ignore the thermal resistance induced by non-cell-stack components such as the tab welding points. For a cell model to accurately predict the effect of how a TMS interacts with a cell while resolving the internal temperature and current, it must be coupled with an anisotropic thermal model that...
captures the non-cell-stack components and realistic thermal boundary conditions. In this work, the development of a 2D thermo-electro model is presented, which can be used to examine the lithium ion cell performance under complex thermal boundary conditions. The model is parameterized for the cell studied in previous work in order to reproduce the effect of tab and surface cooling in realistic scenarios. For example, the thermal resistance caused by the welding of the tabs are carefully calibrated with thermal experiments as described in the Experimental section.

**Model Development**

**Overview.**—The model presented was developed in MATLAB R2017a using Simulink (v8.8) with the Simscape toolbox (v4.1). Figure 1 illustrates the model setup. To reduce the simulation complexity the dimension across the width (W) of the cell was ignored under the assumption that the thermal gradient along this direction is negligible.

The cell was discretized into a number of unit cells and connected together in a 2D network. In this work, the length (L) and thickness (T) plane are simulated. For each unit cell, an ECN model is used to simulate the electrical response of the cell. A thermal ECN model containing the anode, cathode, separator and current collectors are also used. For this approach to work, it was assumed that the cell material and construction are homogenous, therefore it can be modelled with an arbitrary number of identical unit cells.\(^{39}\)

At cell surfaces and tabs, various thermal ECN configurations are used to model different thermal boundary conditions as well as the non-cell-stack components. Figure 1 shows an example of a thermal ECN at the boundary where the cell-tab connection thermal resistance as well as the thermal boundary condition at the electrical tabs are modelled using a thermal ECN model. Various other thermal ECN configurations were applied to model different boundary conditions, such as forced convection, direct liquid cooling, and adiabatic condition.

**Equivalent circuit model.**—Each unit cell is modelled using the equivalent circuit approach, which is commonly used to model the dynamic behavior of lithium ion batteries. As shown in Equation 1, the circuits contain a voltage source \(E_{\text{em}}\), a series resistance \(R_s\) and three more parallel R-C branches connected in series. To accurately model the cell performance over a broad range of operating conditions, three branches of parallel R-C were chosen to produce good accuracy without losing physical meaning. The model parameters are both a function of temperature and SoC, which were implemented as lookup tables that provide the parameter values for the circuit elements. The cell terminal voltage is given by:

\[
V_e = E_e - \sum_{j=1}^{3} U_{k,e} - I_e R_{\text{se}}
\]

where \(V_e\) is the cell terminal voltage, \(I_e\) is the applied current, \(E_e\) is the open circuit voltage (OCV), \(U_k\) is the voltage drop per R-C branch and \(R_{\text{se}}\) is the ohmic resistance.

**Thermal model.**—To get an accurate thermal predication, a 2D thermal model is implemented with careful considerations of the thermal boundary conditions. The cell stack is divided in equally distributed volumes, with 6 nodes along the length and 5 nodes through the thickness of the cell.

Each node is connected to the neighboring nodes through the current collectors as shown in the Figure 1. Within each node, the cell components (anode, cathode, current collectors and separator) are modelled individually with respective thermal conductivities and specific heat capacities. The geometric and thermal properties are defined using an equivalent thermal circuit analogy. The physical dimensions of each component depend on the number of layers contained within each node. Various thermal boundary conditions are implemented with thermal circuits and calibrated with experiments. For example, the thermal resistance caused by the thermal gradient along this direction is negligible.

The cell was discretized into a number of unit cells and connected together in a 2D network. In this work, the length (L) and thickness (T) plane are simulated. For each unit cell, an ECN model is used to simulate the electrical response of the cell. A thermal ECN model containing the anode, cathode, separator and current collectors are also used. For this approach to work, it was assumed that the cell material and construction are homogenous, therefore it can be modelled with an arbitrary number of identical unit cells.\(^{39}\)

**Heat conduction.**—Within each node, a pseudo-2D heat transfer representation is used. Along the length direction, it was assumed that the heat is only transferred via the metallic current collector due to its large thermal conductivity in comparison with the electrodes and separator. However, through the thickness direction the heat must conduct through the anode, separator and cathode. The temperature distribution within each cell element is obtained by solving the transient 1D heat conduction equation (in either the length or thickness direction):

\[
\rho_i C_{p,i} \frac{\partial T}{\partial t} = \frac{\partial}{\partial x} \left( k_{i} \frac{\partial T}{\partial x} \right) + q
\]

where \(\rho_i\) is the density of the component, \(C_{p,i}\) is the specific heat capacity of the component, \(T\) is the temperature of the unit cell, \(k_i\) is the thermal conductivity of the component and \(q\) is the heat source.

**Heat generation.**—Heat generation in Li-ion batteries can be categorized as either reversible or irreversible. The reversible heat generation is due to entropy changes in electrodes during the lithiation and delithiation. The irreversible heat generation is a result of losses due to electronic and ionic transport, as well as reaction currents and other overpotentials. The total heat generation is given by:

\[
q_{\text{gen,e}} = q_{\text{gen,rev,e}} + q_{\text{gen,irrev,e}}
\]

where \(q_{\text{gen,e}}\) is the total heat generation, \(q_{\text{gen,rev,e}}\) is the reversible heat generation and \(q_{\text{gen,irrev,e}}\) is the irreversible heat generation.

The irreversible heat is calculated with \(R_{\text{se}}, R_{\text{casing}}\) and \(I_e\) values in each element. There is a separate lookup table for \(\frac{1}{T_{1/2}}\) for each \(\frac{1}{T_{1/2}}\), along with \(T_1\) and \(I_e\) to calculate the reversible heat. The sum of the heat is then added to each element:

\[
q_{\text{gen,rev,e}} = I_e T_e \left( \frac{dE}{dT} \right)_{\text{SoC}_e} + \sum_{j=1}^{3} R_{e,j} I_e^2 + I_e^2 R_{\text{se}}
\]

where \(I_e\) is the applied current, \(T_e\) is the temperature of the unit cell, \(E\) is the OCV, \(R_{e,j}\) is the resistance of individual R-C branches and \(R_{\text{se}}\) is the ohmic resistance.

**Heat loss to environment.**—There are four surfaces where the heat loss could occur: at the top and bottom of electrode surfaces; and the positive and negative tabs. At each surface, three types of boundary conditions can be applied: (i) Natural or forced convection in ambient; (ii) Direct conduction to liquid-cooled heatsink; (iii) Thermal insulation. Equivalent thermal resistances for each boundary conditions are calculated as shown in Equation 5 and Equation 6. The heat loss due to different thermal boundaries is given in Equation 7.

\[
q_{\text{conv}} = \frac{T_1 - T_2}{R_{\text{total,1-2}}}
\]

Where \(q_{\text{conv}}\) is the heat loss due to convection, \(T_1\) and \(T_2\) are temperature at boundaries and \(R_{\text{total,1-2}}\) are the equivalent thermal resistance.

Total equivalent thermal resistance at the surface and the tab of the unit cell is given by:

\[
R_{\text{total,surf}} = R_{\text{conv}}^* + R_{\text{insulation}}^* + R_{\text{interface}} + R_{\text{casing}}
\]

\[
R_{\text{total,tab}} = R_{\text{conv}}^* + R_{\text{insulation}}^* + R_{\text{interface}} + R_{\text{tab}}
\]

where \(R_{\text{total,surf}}\) is the total thermal resistance at the surface of the unit cell, \(R_{\text{conv}}^*\) is the equivalent thermal resistance associated with convection, \(R_{\text{insulation}}^*\) is the equivalent thermal resistance associated with the insulation material, \(R_{\text{interface}}\) is the equivalent thermal resistance associated with the thermal interface material, \(R_{\text{casing}}\) is the equivalent thermal resistance associated with of the weld point at each tab and \(R_{\text{tab}}\) is the equivalent thermal resistance associated with of the unit cell casing.
Experimental

A 5 Ah high power pouch cell manufactured by Kokam (Model: SLPB11543140H5) was used for this work. The cell contains a graphite anode and a LiMnNiCoO2 (NCM) cathode. According to the specification sheet the electrolyte consists of a solution of LiPF6 in a mixture of organic solvent Ethylene Carbonate (EC) and Ethymethyl Carbonate (EMC). The cell has dimensions of L 140 x W 42 x T 11.4 mm.

For the ECM parameterization experiments, the cell temperature was controlled by forced air convection using a Binder Cooling Incubator (Model: KB23).

For the thermal parameterization and validation experiments, the external temperature was regulated using an ESPEC environmental chamber (Model: BPL-3). The tab cooled and surface cooled cells were placed in bespoke test rigs that used liquid-cooled heatsinks to control the temperature of the different surfaces. The temperature of the coolant is controlled by using an immersion circulator (Model: PC200 manufactured by Thermo Scientific) with a temperature stability of 0.01°C.

Cells were tested using a Maccor (Model: Series 4000) battery cycling system. Temperature measurements were recorded using a Pico Technology data logger (Model: TC-08) with K-type thermocouples.

Model parameterization.—ECM model.—This type of ECN model can be parameterized by using pulse discharge(PD) measurements or Electrochemical Impedance Spectroscopy (EIS). In this work, pulse discharge measurement was used as it does not require more specialist EIS testing equipment. The measurement involves repetitions of a constant current discharge pulse at 5A followed by a resting period of 2 hours. This process starts from 100% SoC and finishes at 0% SoC. The current input and the corresponding voltage response is shown in Figure 2. The measurements were repeated for a range of temperatures (10°C, 20°C, 30°C and 40°C).

For parameter estimation, the ‘layered’ method developed by Jackey et al. was used. The circuit element values were estimated in a piece-wise fashion by calibrating the simulated discharge pulses and the following relaxations at each SoC and temperature with the corresponding measurements. At end of the estimation, 2D look-up tables were generated for each of circuit elements as functions of temperature and SoC, with the exception of Em which was a function of SoC only.

It was assumed that electrode material, separator material and electrolyte filling were uniform throughout cell stack at beginning
of life. This assumption enables the parameters for an individual unit cell to be calculated from the cell parameters. For each unit cell, the resistances and capacitances of each element were calculated according to Equation 8 and Equation 9.

\[
R_{i,e} = R_{i,cell} \cdot N \tag{8}
\]

\[
C_{i,e} = \frac{C_{i,cell}}{N} \tag{9}
\]

Figure 2 shows the parameterization result of the ECN model. Overall, a good fit was achieved for both the relaxation and under load phases of the discharge. Towards the low SoC region, there was an increase in the fitting error due to a combination of highly non-linear behavior of cell and an increasing error in SoC estimation.

The thermal model was carefully parameterized in this work to ensure accurate temperature predictions. The physical parameters of the cell were taken from our previous work, where they were measured for a disassembled cell.

Heat generation.—The irreversible heat generation can be derived from the parameterized equivalent circuit model without additional parameterization. To obtain the reversible heat generation, the entropy change as function of the SoC was measured using the potentiometric method. The method measures the open circuit voltage (OCV) change at equilibrium state as function of temperature change. During the measurement, the cell is rested for 4 hours after each 1/20C discharge in order for it to reach an equilibrium potential. Subsequently, the environmental temperature is changed in increments of 5°C from 15°C to 40°C. At each temperature step, the cell is rested for 4 hours to reach a new thermal and thermodynamic equilibrium. The OCV change due to temperature change is recorded by the potentiostat. This process was repeated every 4% SoC change until the cell was fully discharged. The resulting data was processed as described by Zhang et al.

Thermal parameter.—The parameters for the major components (electrodes, current collector and separator) of the cell were taken from the literature and our previous work, where the thermal parameters of electrodes and separator were measured whilst they are wetted with electrolyte. These parameters are shown in Table I.

In the transient heat test, the cell is placed in the environmental chamber at 20°C for 3 h to reach thermal equilibrium. Eight thermocouples were placed on the cell: six were evenly distributed along the cell center line and two were placed on each tab. The coolant temperature was maintained at 30°C by the immersion water circulator. For each test, a liquid-cooled heat sink is then placed on one of the surfaces (electrode surface, positive tab, or negative tab). The temperatures are monitored until a new thermal equilibrium is established. The calibrated parameters are shown in the Table II.

Results and Discussion

Model validation.—Adiabatic condition validation – drive cycle.—The model was validated against an independent set of experimental data. The US06 drive cycle derived current profile is used as shown in Figure 3a. The experiment was conducted in a near-adiabatic condition, where the entire cell was covered by the thermal insulation.

![Figure 3. Model validation result against US06 drive cycle: (a) Input current, (b) Terminal voltage, (c) Cell temperature.](image-url)
The same thermal condition was implemented in the model. This thermal boundary was chosen to minimize the internal thermal gradient. In Figure 3c, the temperature measured at the center of the cell surface is compared with the simulation. The simulation again showed good agreement with the experiment. The peak temperature error is under 0.2 °C, while the maximum error is under 0.6 °C. This result indicates the overall cell stack heat capacity matches well with the experimental cell. Figure 3b shows the voltage from the simulation and experiment results. The simulation showed good agreement with the experiment results under non-isothermal condition.

**Under thermal management – thermal distribution.**—In this section, the effect of the thermal gradient was introduced and the simulated temperature distribution was compared against the experiment. Two validation experiments were conducted: 1. Tab cooling; 2. Single-side surface cooling. In each test, the temperature at the designated surfaces was controlled at 20 °C throughout. The temperature distribution was measured at 6 linearly spaced thermocouples across the cell surface as shown in Figure 4. The cell was allowed to reach thermal equilibrium inside the environmental chamber with the heat sink applied to the corresponding surfaces. The cell was then discharged at a current rate of 6C (30A) from 4.2 V to 2.7 V, followed by 2C (10A) charging.

The measured temperature at different locations of the surface-cooled cell is compared against the model prediction in Figure 4a. The model predicted the correct thermal distribution along the measured points, where the temperature is higher at middle (S3 and S4) and lower toward each tab (S1 and S6). Overall, there is good agreement between simulation and experiment during the discharge phase. The error between measurements and simulation is more significant during the subsequent charging phase with a maximum difference of 2.8 °C. Figure 4b shows the result for the tab cooled cell where both tabs were controlled at 20 °C. Experiments indicate that the cell was hotter in the middle (position S3 and S4) and colder toward the tabs (position S1 and S6). The maximum error at S3 is ±2.3 °C between the measurement and simulation. The temperature transient during both discharge and charge follows the measurements very closely, where error is under −0.5 °C. Overall, the model produced acceptable quantitative prediction of cell surface temperature at various locations under both cooling conditions. Based on this validation, we assume that this model is suitable for further investigating the internal states of the cell under operating conditions.

**Internal temperature, current, and SoC distributions.**—Figure 5 shows the simulation result comparing the tab and surface cooled cells’ internal states during a 6C (30A) discharge. Temperature, current, and SoC distributions were plotted at four snapshots during the discharge. At each time step, the distributions are normalized by the average of the corresponding quantity. For example, at t = 0s the plotted temperature is $T_{i,j}(t=0) = \frac{T_{\text{avg},i,j}(t=0)}{T_{\text{avg}}}$, where i and j are coordinates of simulated nodes. The normalization allows a better visualization of the inhomogeneity within the simulated domain.

The simulation domain is shown in Figure 5a as described in the Overview section. The simulated nodes are plotted as a black marker in each plot and the distribution map is created by linearly interpolating between nodes. For the surface cooled cell, the surface at Thickness = 5 mm is controlled at 20 °C and the starting temperature is 20 °C. For the tab cooled cell, the tabs were held at 20 °C during discharge and the starting temperature is 20 °C. For both cases, the rest of the surfaces are thermally insulated.

Figure 5b shows the predicted temperature distributions during the discharge period for both the surface- and the tab-cooled cells. For the tab cooled cell, the average cell temperature increases from 20 °C to 40 °C by the end of the discharge. At t = 185s, the maximum temperature is 0 °C above the average and the minimum temperature is 0.6 °C below. The maximum temperature is located toward the center of the cell, while the minimum is at the negative tab end. At t = 560s, the maximum is 0.7 °C above and the minimum is 1.2 °C below the average temperature, with maximum temperature gradient of 1.9 °C. In comparison, the surface cooled cell has a lower average temperature during the discharge; 3°C colder at t = 180s and 8°C colder at t = 550s. However, the surface cooled cell experienced much greater thermal inhomogeneity; 11°C of maximum temperature difference at t = 180 s compared to 1.9 °C for the tab cooled cell. The average thermal gradient is approximately 1 °C/mm for the surface cooled along thickness direction. For the tab cooled cell, the average thermal gradient is only 0.03 °C/mm along the length direction.

Figure 5c shows the current distribution during the discharge period. Because of the thermal inhomogeneity, the current generated at each node is not uniform. During the constant current discharge, the average current is controlled at 6C (30A). For the surface cooled cell, the hotter part of the cell discharged faster at the beginning. The top layers (thickness = 5 mm) discharged at rate of approximately 6.3C till t = 365s. The colder part of cell discharged at a much slower rate, approximately 5.3C at t = 180 s and 4.9C at t = 365 s. The current difference is a direct consequence of the thermal gradient: the hotter part of the cell has lower impedance which leads to more current being generated, and vice versa for the colder part.

The colder cell region starts to contribute more current toward end of discharge, where the bottom layer (thickness = 5 mm) discharges at a rate of 7.9 C, 1.9 C higher than the average. The hotter part discharges 1C below the average. This reversion of current distribution is caused by the lower SoC of the hotter region, which had been discharging faster than the colder part of the cell, as can be seen in Figure 5d at t = 365s. Lower SoC corresponds to higher overpotentials and a lower OCV, which toward the end of discharge, the effect of the higher temperature can no longer offset, hence the colder cell region will start to provide more current.

For the tab cooled cell, the current is distributed much more uniformly. At t = 185s, the maximum discharge rate is 6.1C at the center and the minimum is 5.9C toward the negative tab. At the end of the discharge, the maximum discharge rate is 6.3C at the center and minimum 5.4C toward negative tab. The discharge rate inhomogeneity (difference between maximum and minimum) increased from 0.2C to 0.7C from t = 185s to t = 560s. This is caused by the thermal gradient built up. However, the current distribution trend is unchanged during the discharge.

Comparing the surface-cooled and tab-cooled cells, the surface cooled cell again showed more inhomogeneous behavior. The initial temperature gradient caused by the external cooling source led to unequal current flow, where more current is flowing through the hotter part of the cell. The above-average current in this part of the cell then leads to more heat generation that will further exacerbate the thermal gradient. This positive feedback loop continues until the ‘hotter’ part of the cell can no longer sustain the higher current rate. Subsequently, the ‘colder’ part starts to provide more current and leads to higher-than-demanded current rate. For tab cooled cell, this effect happens only on a much smaller scale as the thermal gradient is significantly smaller.

Figure 5d shows the SoC distribution during the discharge period. As the result of the temperature and current positive feedback loop, the surface cooled cell is discharged unequally. At t = 180s, the maximum SoC is 1.5% above the average and the minimum SoC is 0.7% below. This difference increased to the maximum of 5.5% above average and the minimum of 2.6% below at t = 365s. Towards the end of discharge, this difference became less with the maximum of 4.9% above average and the minimum of 1.6% below.

For the tab cooled cell, the SoC distribution is more homogeneous. The maximum SoC is at t = 370 s, with the different between maximum and minimum up to 1.6%. The maximum is 1% above the average and the minimum is 0.6% below. Similarly, the difference reduced toward the end of discharge, with the maximum difference reduced to 0.6%.

In comparison, the surface cooled method induced a much bigger SoC gradient during discharge. This also led cell to reach the end of discharge quicker, therefore reproducing the experimentally observed reduction in usable capacity reported in our previous work.17
Effect of operating current and temperature.—The thermal boundary condition clearly has a significant effect on cell performance. In this section, the severity of this effect is assessed across a range of operating conditions. The effect of the discharge current and the temperature of the heat sink is investigated with different discharge currents under constant boundary temperature of 20°C, and a range of different boundary temperatures at a constant discharge rate of 6C.

In each case, four performance indicators are chosen to assess the performance of the system, these indicators are: temperature, heat flux extracted, current distribution, and SoC at the end of the discharge. For temperature, the cell average, maximum and minimum temperature at the end of the discharge are plotted. For heat flux, the average and the peak heat extracted during the entire discharge are plotted. For current, the maximum and minimum current in the cell during the entire discharge are plotted. For SoC, the average remaining SoC of the cell, the maximum and the minimum SoC within cell at the end of the discharge are plotted.

Figure 6 compares the performance of surface and tab cooled cell performance at different discharge rate. Figure 6a shows temperature at the end discharge. For the surface cooled cell, the average end of discharge temperature rises from 21°C to 32°C as the discharge current increases from 1C to 6C. The temperature deviation from the average has also increased as the rate increases. The maximum temperature difference increased from less than 1°C at 1C to more than 7°C at 6C. The major thermal gradient is still in the thickness direction. For the tab cooled cell, the average temperature is higher, raising from 23°C to 40°C. The maximum temperature difference is less than 0.1°C at 1C and less than 1°C at 6C. In comparison, the tab cooled cell was on average hotter compared to the surface cooled cell with a difference of 2.5°C at 1C and 8°C at 6C. However, the temperature gradient for the tab cooled cell is significantly smaller in comparison.

Figure 6b shows the average and maximum heat flux extracted by the cooling system during the discharge. For the surface cooled cell, the average heat flux increased from 0.5 W to 8.5 W from 1C to 6C. The maximum increased from 1.3 W to 14.5 W. For the tab cooled cell, the average heat flux increased from 0.5 W to 4.4 W from 1C to 6C. The maximum heat flux is 1.5W at 1C and 7.5W at 6C. In comparison, tab cooling is much less capable of removing the heat from the cell above a discharge rate of 2C. At higher current rates the maximum heat removal rate becomes the limiting factor for tab cooling compared to the surface cooled cell. At 6C, tab cooling method was only being able to remove half of the heat compared to
surface cooling, and this was only because the cell was significantly hotter. At lower currents < 2C, the performances of the two cooling methods are similar. This result would be different for different cells.

The consequence of the thermal gradient is shown in Figures 6c and 6d, where 6c shows the current inhomogeneity during the discharge and the (d) shows the SoC inhomogeneity at the end of discharge. For the surface cooled cell, the maximum deviation from the demanded current is approximately 0 at 1C rate and 2C at 6C rate. This means part of the surface cooled cell could be discharged at 8C during a 6C discharge. This deviation could lead to inhomogeneous degradation where the cell could be degraded much faster at the region with a higher discharge rate. The cumulative consequence is shown in the

Figure 5. Cell state during 6C discharge. [a] Temperature distribution, [b] Current distribution, [c] State of charge distribution.
Across different starting temperatures ($T_{\text{start}}$). Figure 7 shows the performance comparison, where temperature is the starting temperature of the cell and the ambient temperature. The applied cooling system temperature is 20°C in all cases, i.e., heat sink. Figure 7a shows the temperature of the cell at the end of discharge. The surface cooled cell, the average temperature increases from 25°C to 43°C as $T_{\text{start}}$ increases from 10°C to 35°C. The temperature gradient within the cell decreases as the operating temperature increases, and the maximum difference is 9°C at $T_{\text{start}} = 10$°C and the difference is 5°C at $T_{\text{start}} = 35$°C.

For the tab cooled cell, the average temperature increases from 35°C to 50°C as the $T_{\text{start}}$ increases. However, the temperature gradient is negligible in all cases. In comparison, the tab cooled cell is hotter at the end of discharge, the difference is 10°C at $T_{\text{start}} = 10$°C and 7°C at $T_{\text{start}} = 35$°C. Tab cooling is able to keep the cell at uniform temperatures in all cases.

Figure 7b shows the heat flux during the discharge. For both cell, the average heat flux during discharge reduces as $T_{\text{start}}$ increases. Surface cooling removes 11 W on average at $T_{\text{start}} = 10$°C and 5 W on average at $T_{\text{start}} = 35$°C. In contrast, the tab cooled cell only removes 6 W at $T_{\text{start}} = 10$°C and 3 W at $T_{\text{start}} = 35$°C. The tab cooled cell again is limited by the heat removal rate. This is despite the heat needing to be removed decreasing as operating temperature increases, which is due to the reduction in overpotential from faster kinetics and transport at higher temperatures.

Figure 7c shows the average and maximum heat flux extracted by the cooling systems during discharge. It appears there is little correlation between peak current and operating temperature. However, a closer inspection of current as a function of time in each case shows that the current is much more homogeneous at higher temperature. The current flow in each layer/section of the cell is much more uniform at higher temperature, while the peak and minimum are similar in each case. This effect is clearly demonstrated in Figure 7d, where the plot shows the maximum SoC deviation from the average at end of discharge. For the surface cooled cell, the average SoC remaining at the end of discharge is reduced from 12% at $T_{\text{start}} = 10$°C to 5% at $T_{\text{start}} = 35$°C. This is expected as higher temperature reduces cell impedance, therefore the cutoff voltage is not exceeded until much later during the discharge. The maximum SoC gradient is over 11% at $T_{\text{start}} = 10$°C. It indicates the cell has not been discharged uniformly, where the section with the lowest remaining SoC has discharged at higher current than the average demanded. This gradient reduces to 0.5% at $T_{\text{start}} = 35$°C.

For the tab cooled cell, the remaining SoC only reduced from 7% to 5% as at $T_{\text{start}}$ increased from 10°C to 35°C. It remained very uniform across cell sections at all temperature.

To sum up, tab cooling is limited by the rate of heat removal at all temperature ranges. However, tab cooling provides better thermal uniformity, therefore more uniform current and SoC distributions. These factors increase usable capacity during discharge.

Conclusions

A 2D electro-thermal model for Li-ion cell has been developed to investigate the effect of the thermal gradient on lithium ion cell performance. The model consists of a fully coupled electrical model, heat generation model, and a detailed heat transfer model in 2D.

An ECN model is used to model the unit cell in a 2D space. The heat generation model takes account into both reversible and irreversible heating. A thermal ECN model is used to model each unit cell including major cell components, such as anode, cathode, separator and current collectors. The non-cell-stack component such as the tab weld were also modelled to ensure accurate internal temperature prediction.

The thermal model was carefully parameterized with transient thermal measurements. The full model was validated with drive cycle tests. The model thermal distribution prediction was validated against experiments under different thermal boundary conditions.

The model is used to assess the tab cooling strategy in comparison to the surface cooling strategy. At high discharge of 6°C, the surface cooling is able to maintain the cell at a lower average temperature at the expense of creating a significant larger thermal gradient. While
the tab cooled cell is operating at a higher average temperature, the thermal gradient within the active regions of the cell is minimal. The large thermal gradient induced by the surface cooling method led to significant current and SoC inhomogeneity. These effects lead to a reduction in usable capacity, correlating well with our previous work,\(^1\) where surface cooling is observed to cause significantly more degradation in comparison to tab cooling.

The performance of the two cooling strategies are assessed across a range of temperature and discharge currents. The performance was assessed based on the average temperature, temperature gradient, ability to extract heat, current and SoC homogeneity. At high current rates, the tab cooling maintained much better thermal uniformity which led to homogenous current and SoC distribution. This allowed more capacity to be extracted. However, the tab cooling system is limited by its ability to extract heat and as a result, the cell operates at a higher temperature. At low temperature, the similar effect is also observed, where surface cooling also induces large thermal gradient and causes significant inhomogeneity.

In conclusion, correctly modeling the thermal boundary conditions around the tab are critical to predict the performance of tab cooling and ignoring this region in a model will lead to incorrect results. For tabs held at 20 °C it was found that the coldest region of the active layers in the cell nearest the negative tab was 38.8 °C and the hottest region in the middle of the cell was 40.7 °C, indicating a thermal gradient of just 1.9 °C within the active regions of the cell but a thermal gradient of 18.8 °C between the active regions and the tab. Clearly the thermal resistance between the current collectors and the tab. Clearly the thermal resistance between the current collectors and the tab represents a significant heat transfer bottleneck and efforts to improve this could have highly significant positive impacts on the performance of lithium ion batteries considering the other advantages of tab cooling.

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