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# Accepted Manuscript

Research Paper

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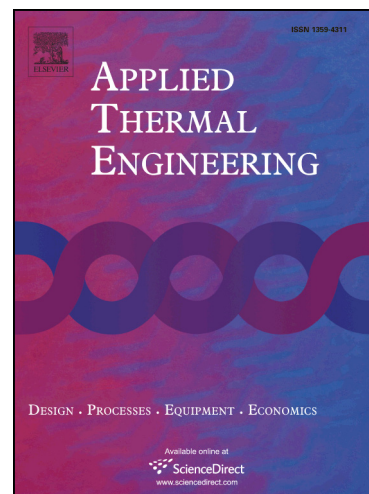
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## A multilayer electro-thermal model of pouch battery during normal discharge and internal short circuit process

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Highlight:

2D network equivalent circuit considers the interplay of cell units.

The temperature non-uniformity  $\Phi$  of multilayer model is bigger than that of lumped model

The temperature non-uniformity is quantified and the reason of non-uniformity is analyzed.

Increasing the thermal conductivity of the separator can effectively relieve the heat spot effect of ISC

Abstract: As the electrical and thermal characteristic will affect the batteries' safety, performance, calendar life and capacity fading, an electro-thermal coupled model for pouch battery LiFePO<sub>4</sub>/C is developed in normal discharge and internal short circuit process. The battery is discretized into many cell elements which are united as a 2D network equivalent circuit. The electro-thermal model is solved with finite difference method. Non-uniformity of current distribution and temperature distribution is simulated and the result is validated with experiment data at various discharge rates. Comparison of the lumped model and the multilayer structure model shows that the temperature non-uniformity  $\Phi$  of multilayer model is bigger than that of lumped model and shows more precise. The temperature non-uniformity is quantified and the reason of non-uniformity is analyzed. The electro-thermal model can also be used to guide the safety design of battery. The temperature of the ISC element near tabs is the highest because the equivalent resistance of the external circuit (not including the ISC element) is the smallest when the resistance of cell units is small. It is found that increasing the thermal conductivity of integrated layer can effectively relieve the heat spot effect of ISC.

Keywords: Lithium ion battery; Electro-thermal coupled model; temperature distribution; internal short circuit

## 1. Introduction

Lithium ion batteries with its high specific energy, high theoretical capacity and good cycle-life are gaining popularity for Electric Vehicles (EVs) and Hybrid Electric Vehicles (HEVs) applications. However, a series of fire accidents indicate that thermal safety is still challenge to be overcome [1]. Thermal issue such as uneven temperature profile of batteries or high heat generation will make a great difference to its safety, performance, calendar life and capacity fading[2, 3]. Little non-uniformity of temperature can lead to great inconsistency of batteries. The inconsistency of batteries can do great harm to the performance of battery packs. Moreover, thermal runaway induced by internal short circuit (ISC) may lead to fire explosion. Therefore, thermal management system (TMS), which aims to optimize the performance by controlling the temperature distribution and to prevent the thermal runaway by controlling the heat transfer, is required.

Many literatures have investigated the thermal behavior of batteries and tried to predict the heat generation with different methods. A general heat source model [4] approved by D. Bernardi, was widespread used. The model divided the heat source into several terms including heat from reaction of electrochemical, heat from mixing, heat from phase change and joule heating. Other heat source models developed including electrochemical-thermal model and electro-thermal model. Electrochemical-thermal model based on porous electrode theory and Fick's law is used to describe electrochemical process and the mechanism of change of heat generation [5-10]. Electrochemical model is precise but it depends much on the parameter which is quite hard to measure. Also it is

hard to solve the complicated partial differential equations of the electrochemical model. The precision will decline in large format battery with 1D electrochemical model because it neglects the interplay of other cell elements in X-axis and Y-axis (figure 1). However, a 2D network equivalent circuit method can take it into account. Moreover, electro-thermal model based on 2D network equivalent circuit is simple and the parameter is easy to measure [1, 11-14]. It uses impedances or run time based model to represent the characteristics of the batteries [13].

LiFePO<sub>4</sub> electrode with olivine structure is a promising candidate electrode material because of its low price and high safety performance. Ahmadou Samba [15] developed a 2D thermal model to predict the temperature distribution but it did not consider the current distribution. M. Xu [9] used a pseudo 3D electrochemical-thermal model to describe the thermal behavior but it ignored the layer structure of pouch battery. C. Veth [14] used 3D electro-thermal model to simulate the current density and temperature distribution but the cause of non uniform distribution was not fully discussed and relation between temperature and property was implicit. Li Teng [16] separated the battery into several layers but the current density was implicit and the effect of multi-layer was not discussed.

Besides the prediction of temperature distribution, many literatures and patents have tried to predict and detect the internal short circuit (ISC) as it will generate a great amount of heat in a short time [17]. If the heat is not dissipated efficiently, the battery temperature will exceed a critical point, above which the electrolyte, active materials, and SEI (solid electrolyte interface) film will decompose, and eventually thermal runaway and even fire will take place [17]. Nail penetration experiment is often used to study the performance of ISC [17-18]. However, the fact that ISC probably occurs in a single cell unit (figure 1) is different from nail penetration

experiment in which the whole battery is penetrated. 1D Electrochemical model is excellent to study the mechanism of ISC [18-19] but it neglects the interplay between the short spot and the other parts of battery.

The pouch battery can be divided into many layers as illustrated in figure 1. Cathodes and anodes are coated in aluminum and copper foils. The property of each layer is different and the calculation of temperature distribution is not precise when we ignore the layer structure and the current distribution.

In this work, a multi-layer 3D thermal model is developed to simulate the temperature distribution of the battery while a 2D network equivalent circuit is developed to calculate the heat source. Detailed study of the thermal behavior of the battery during normal discharge and ISC process is discussed.

## 2. Model

A pseudo 3D electro-thermal model for a single  $\text{LiFePO}_4$ /graphite battery is developed based on a 2D network equivalent circuit and energy conservation. The nominal capacity is 20 Ah. The size of the pouch battery is 170 mm  $\times$  230 mm  $\times$  7 mm (width  $\times$  length  $\times$  thickness). The thickness of positive current collector (aluminum foil) is 20  $\mu\text{m}$  while the thickness of negative collector (copper foil) is 14  $\mu\text{m}$ . As illustrated in figure 1, we integrate positive electrode, separator and negative electrode as one integrated layer because the thermal property of separator and active material is much different from the current collector layer. The thickness of integrated layer is 140  $\mu\text{m}$ . Cathodes and anodes are coated in aluminum and copper foils. We found that the battery contains 17 aluminum foil sheets, 16 copper foil sheets and 32 integration sheets after the

disassembly. As one cell unit is combined with one aluminum foil layer, one integration layer and copper foil layer, one pouch battery is combined with 32 cell units connected in parallel. That means a battery can be assumed as a “cell unit module”.

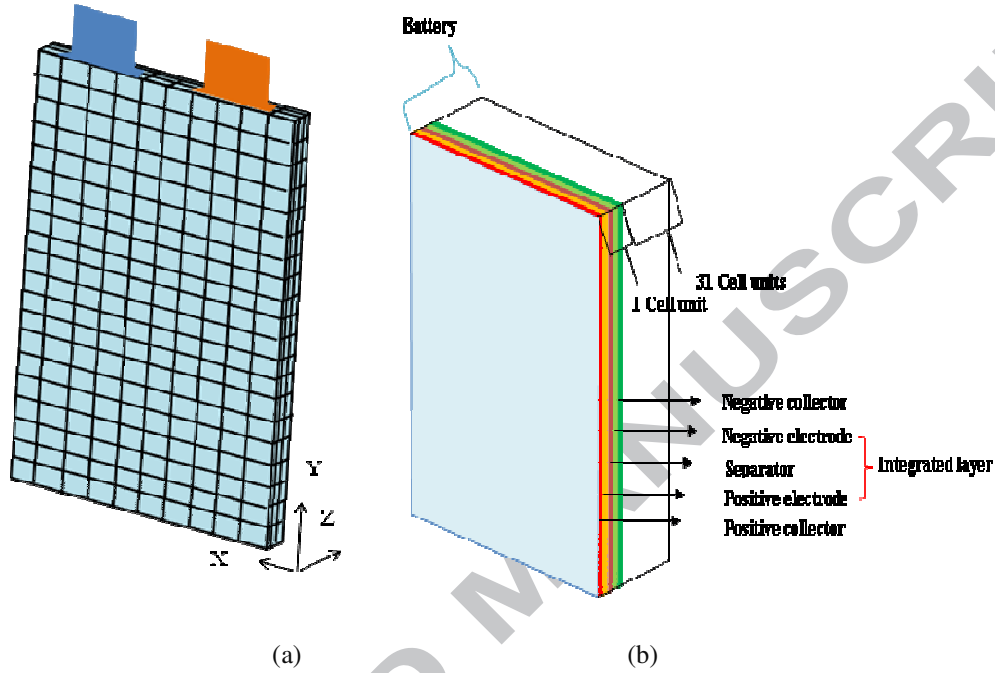


Figure 1 Layer structure of pouch battery (the ratio of size is adjusted for better illustration)

## 2.1 Thermal model

For a pouch battery, the heat transfer should follow the energy conservation law:

$$\rho c \frac{\partial T}{\partial t} = \lambda \nabla^2 T + q \quad (1)$$

where  $\rho$  is the density,  $c$  is heat capacity,  $\lambda$  is the heat conductivity,  $T$  is the temperature,  $t$  is time and

$q$  is the heat source.  $q$  is the summation of all the heat generations. We neglect the heat of phase

change so that the heat generation includes there terms:

$$q = q_J + q_p + q_r \quad (2)$$

where  $q_J$  is the joule heat,  $q_p$  is the irreversible electrochemical reaction heat and  $q_r$  is the reversible



entropic heat. If the electrochemical reaction is ideally reversible, the thermodynamic equation under a constant temperature  $T$  and constant pressure yields the following relation[20]:

$$\Delta G = \Delta H - T\Delta S \quad (3)$$

where the equation indicates the residual energy of reaction enthalpy change  $\Delta H$  and electrical work ( $\Delta G = -nFE_e$ ) is released as heat energy  $q_r$ . And the reversible heat is described as:

$$q_r = T\Delta S \frac{I}{nF} \quad (4)$$

$$\Delta S = \frac{\partial \Delta G}{\partial T} = nF \frac{\partial E_e}{\partial T} \quad (5)$$

where  $I$  is the charge/discharge current,  $F$  is Faraday constant,  $E_e$  is the cell potential for open-circuit and  $n$  is the charge number pertaining to the reaction. We can get the value of  $\partial E_e / \partial T$  from empirical equation or through the experimental data[11, 21]. The acquisition of  $q_j$  and  $q_p$  is described in electrical model in the next section.

Noting the convective heat transfer and radiation heat transfer on the boundary, we get equation (6)

$$\begin{aligned} q_s &= (h_{conv} + h_{rad})(T - T_{am}) \\ h_{rad} &= \varepsilon\sigma(T^2 + T_{am}^2)(T + T_{am}) \end{aligned} \quad (6)$$

where  $h_{conv}$  is the convective heat transfer coefficient,  $h_{rad}$  is the radiation heat transfer coefficient,  $T_{am}$  is the ambient temperature,  $\varepsilon$  is surface emissivity, and  $\sigma$  is the Stefan-Boltzmann constant.

The tabs are considered and the heat is conducted to the electrodes according to Fourier law of heat conduction.

## 2.2 Electrical model

A 2D network equivalent circuit is used in order to take the interplay of different cell

elements into consideration. The interplay is overlooked in 1D electrochemical model. One pouch battery is combined with 32 cell units connected in parallel. One cell unit can be discretized into many volume elements. Each volume element is regarded as a small cell element. When the volume element is small enough, it can be assumed that the temperature of that element is uniform. On the other hand, the volume element could not be too small because it should be large enough to represent the average value of a cell element. Therefore, we divide one cell unit into  $20 \times 10 = 200$  cell elements and one pouch battery is connected by  $20 \times 10 \times 32 = 6400$  cell elements as illustrated in figure 1. The equivalent circuit[22] model of a complete battery is spatialized to 2D network model as illustrated in figure 2. According to the double electrode layer theory, the diffusion layer of LiFePO<sub>4</sub> battery is obvious enough to use a second order model[23]. The second order model contains  $V_{OCV}$  (open circuit voltage), polarization resistance  $R_p$  (including electrochemical polarization resistance  $R_{ep}$  and concentration polarization resistance  $R_{cp}$ ), polarization capacitances ( $C_{ep}$  and  $C_{cp}$ ), and ohmic resistances ( $R_0$ ,  $R_{ta,A}$ ,  $R_{ax,A}$ ,  $R_{ta,C}$  and  $R_{ax,C}$ ). The parameters are depended on the temperature, SOC (state of charge) and rate of charge/discharge. The  $V_{OCV}$  of each cell element is almost the same to the  $V_{OCV}$  of the battery because the equilibrium potential is determined by the material of electrode/ electrolyte, concentration and temperature according to the Nernst equation. Eight resistances on the top and the bottom denote the positive current collector and negative current collector. The resistances on the current collector of each cell element are connected to the corresponding resistances of the adjoining cell element but the polarization resistances of each cell element are relatively independent from each other. All the cell elements in one cell unit are connected as a network. The resistances on the current collector are determined by the geometry and the material.

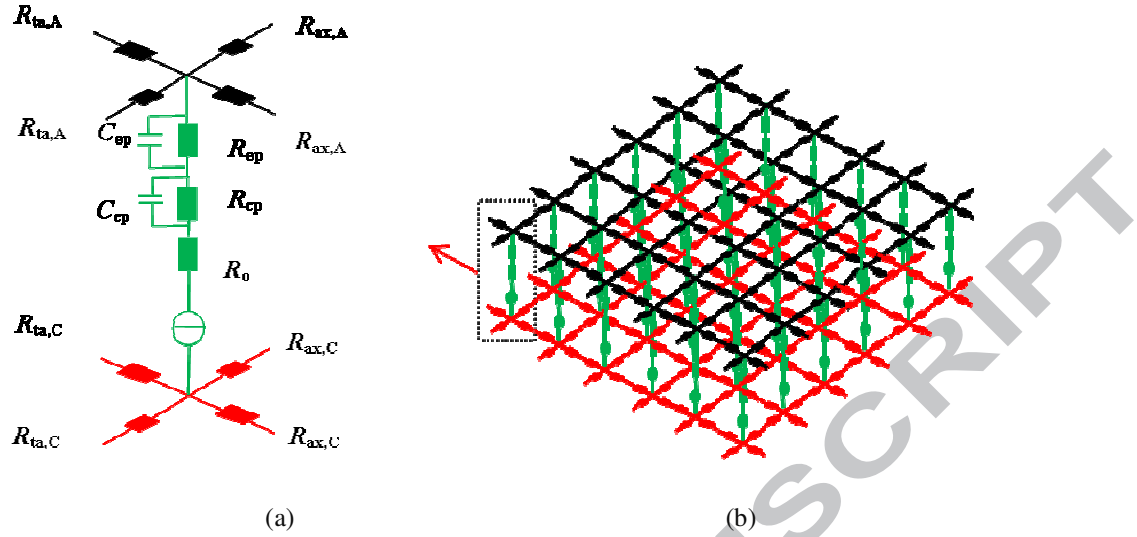


Figure 2 Illustration of the cell element equivalent circuit (a) and 2D network equivalent circuit.

Polarization capacitances are not illustrated for better visualization (b)

One pouch battery is connected by 32 cell units in parallel and it is assumed that the current between different cell units (not cell elements) is independent. It is rational in normal discharge process because the resistance and the current distribution in each cell unit are almost the same. We suppose that the current is divided into 32 parts equally and each part will flow into one cell unit. Every current collector except two at the edge will undertake the currents generated by integrated layers on both sides. Then, we just need to focus on the grid circuit combined with  $20 \times 10 = 200$  cell elements in one cell unit. What we concern about is the current in each circuit branch. Based on the Kirchhoff's current law, the sum of currents flowing into a node is equal to the sum of currents flowing out of that node.

$$\sum_{k=1}^m I_k(T, SOC) = 0 \quad (7)$$

where the  $I_k$  is the branch current flowing in or out of the node. According to the Kirchhoff's voltage law, the directed sum of the electrical potential differences (voltage) in a closed loop is zero.

$$\sum_{k=1}^m V_k(T, SOC) = 0 \quad (8)$$

where  $m$  is the total number of electric elements in a closed loop and  $V_k$  is the voltage of the electric element (polarization resistances, resistances on the current collector and the power source). In discharge process, we acquire equation (9) from the second order model.

$$\begin{aligned} V_t(T, SOC) &= V_{ocv}(T, SOC) - I_k(T, SOC) \cdot R_o(T, SOC) - V_{ep}(T, SOC) - V_{cp}(T, SOC) \\ V_{ep}(T, SOC) &= I_k(T, SOC) R_{ep}(T, SOC) \cdot (1 - e^{-t/\tau_{ep}}) \\ V_{cp}(T, SOC) &= I_k(T, SOC) R_{cp}(T, SOC) \cdot (1 - e^{-t/\tau_{cp}}) \end{aligned} \quad (9)$$

where the  $\tau_{ep}$  and  $\tau_{cp}$  are the time constant which can be measure from experiment[23]. They almost remind unchanged in different temperature and SOC.

From all independent nodes and closed loop in the network, independent closed equations are list based on equations (7-9). We acquire the current in each branch after solving the equations. Therefore, for each cell element we can calculate the irreversible electrochemical reaction heat  $q_p$  and joule heat  $q_j$ .

$$q_j + q_p = \sum I_k^2(T, SOC) \cdot R_k(T, SOC) \quad (10)$$

where  $R_k$  ( $R_{ep}$ ,  $R_{cp}$ ,  $R_0$ ,  $R_{ta,A}$ ,  $R_{ax,A}$ ,  $R_{ta,C}$  and  $R_{ax,C}$ ) is the corresponding resistance illustrated in figure 2 and  $I_k$  is the branch current flowing through  $R_k$ . In electrical model, temperature and SOC are input while the sum of irreversible electrochemical reaction heat and joule heat is output. The calculation of SOC is based on Ampere-Hour integral method.

## 2.3 Internal short circuit

As mentioned before, complex electrochemical reaction will occur and lead to thermal runaway when the battery temperature exceeds a critical value[24]. The SEI film will melt and the electrolyte will decompose around 120 °C. That will lead to further reaction of the lithiated

graphite and the electrolyte [17, 24]. Therefore, we just focus on the ISC process before the temperature exceeds 150 °C. Major assumptions are described: no venting, no combustion, structurally intact; short remaining the same [25]. We suppose that the current is not limited by the mass transportation. In the process of ISC, the ISC resistance and the  $V_{OCV}$  will diminish [26]. In the electrical model, we replace the resistance  $R_{cp}$ ,  $R_{ep}$  and  $R_{oh}$  (figure 2) with short resistance  $R_{short}$  at the short cell element.

We study the feature of anode-cathode short and anode-aluminum short of single cell unit. The anode-aluminum short is considered by some studies to be more dangerous as the resistance at the short spot is relatively low and lack of good heat dissipation [19]. We assume the resistance of an-al short is around 30m $\Omega$  while the resistance of an-ca short is 300m $\Omega$  according to the data of reference [27].

In normal discharge process, current in different cell units is independent but now it is dependent in internal short circuit process. The intact cell units are regarded as power source with internal resistance. Heat generation from ISC contains the heat from discharge and the Joule heat at short spot.

## 2.4 Parameter

The battery is divided into many layers and the property of each layer is different. It is assumed that the thermal property including specific heat capacity, thermal conductivity and density does not change with temperature. They are given in table 1.

Table 1 Thermal property of different layers of battery

	specific heat capacity /(J·kg <sup>-1</sup> ·K <sup>-1</sup> )	thermal conductivity /(W·m <sup>-1</sup> ·K <sup>-1</sup> )	density /(kg·m <sup>-3</sup> )
Aluminum foil layer	880	200	2700

Integrated layer[28]	2738	0.11	1615
Copper layer	384	375	8920

For the parameters in electrical model, the  $V_{OCV}$  (open circuit voltage) is measured with experiment. Resistance of battery includes polarization resistances ( $R_{ep}$  and  $R_{cp}$ ) and total ohmic resistances ( $R_{ohm}$ ). They are measured by chronopotentiometry method. The potential response is shown in figure 3 (a). During the discharging process, we cut down the current and measure the voltage of the battery at that transient state. The response of ohmic resistance is very fast and the response of polarization is lagging. They are given in equations:

$$R_{ohm} = \frac{U_a - U_b}{I}$$

$$R_{ep} + R_{cp} = \frac{U_a - U_c}{I} - \frac{U_a - U_b}{I} \quad (11)$$

The  $U_a$ ,  $U_b$ , and  $U_c$  are shown in figure 3(a). The experiment data is illustrated in figure 3 (b). Noticing that the ohmic resistance remains nearly unchanged, it means the result is rational. Besides the method we used, it is reasonable to measure the resistances with the electrochemical impedance spectroscopy (EIS).

Of course, the resistances measured from the experiment belong to the whole battery. Therefore, they should be transferred to the cell elements' resistances based on the aforementioned electrical model. Collector resistances are calculated based on material and geometry. The ratio of  $R_{po}$ ,  $R_c$  and  $R_{ohm}$  is fixed and the ratio value is based on the measure. Anyway, in constant-current discharge mode, the ratio is not so important in thermal calculation because the sum of these resistances is nearly invariable after 100 s discharging according to equations (9).

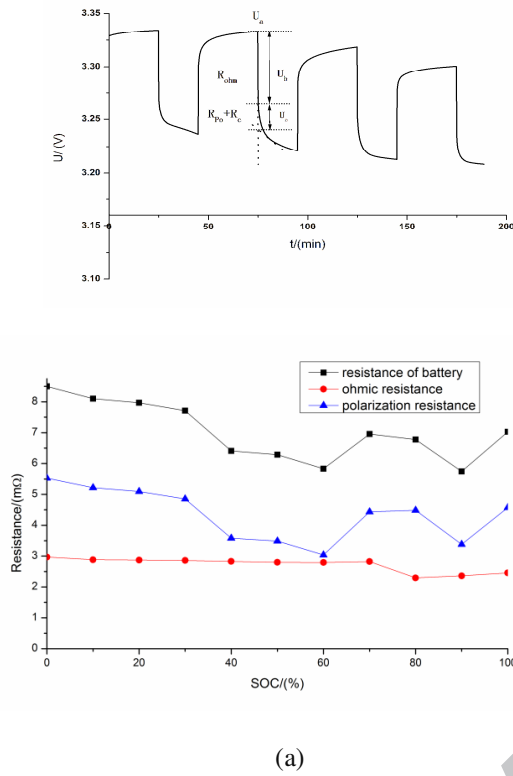


Figure 3 The potential response of battery (a) and resistance of battery (b) measured with chronopotentiometry method

## 2.5 Solution of equations

As mentioned before, a pouch battery contains 17 aluminum foil layers, 16 copper foil layers and 32 integration layers. A cell unit is discretized into  $20 \times 10$  cell elements in electrical model. Therefore, in the thermal model the pouch battery is discretized into  $20 \times 10 \times (17+16+33) = 13200$  volume elements correspondingly. The control equation and the boundary condition equations are described in the aforementioned thermal model while the heat source term can be acquired from electrical model. Then, the finite difference method is used to solve the partial differential equations.

In the thermal model solution, the number of the equations is large and the nonzero element

in the coefficient matrix is sparse. Therefore we use Gauss–Seidel method for the iterations. In the electrical model, we use Gaussian elimination method to solve the equations. We note that the change of temperature from thermal model would influence the parameter of electrical model while the change of electrical model would influence the generation of the thermal model. In order to decouple, the temperature used in the electrical model comes from the thermal model at the last moment. The flowchart can be seen in figure 4. The calculation procedure is written in C language by ourselves and run in Linux operation system.

Result of finer grid ( $30 \times 20 \times (17+16+32) = 39000$  volume elements) is compared with the coarse grid ( $20 \times 10 \times (17+16+32) = 13000$  volume elements). The difference of simulated temperature is below 3%. That means the grid is fine enough with  $20 \times 10 \times (17 + 16 + 32) = 13000$  volume elements.

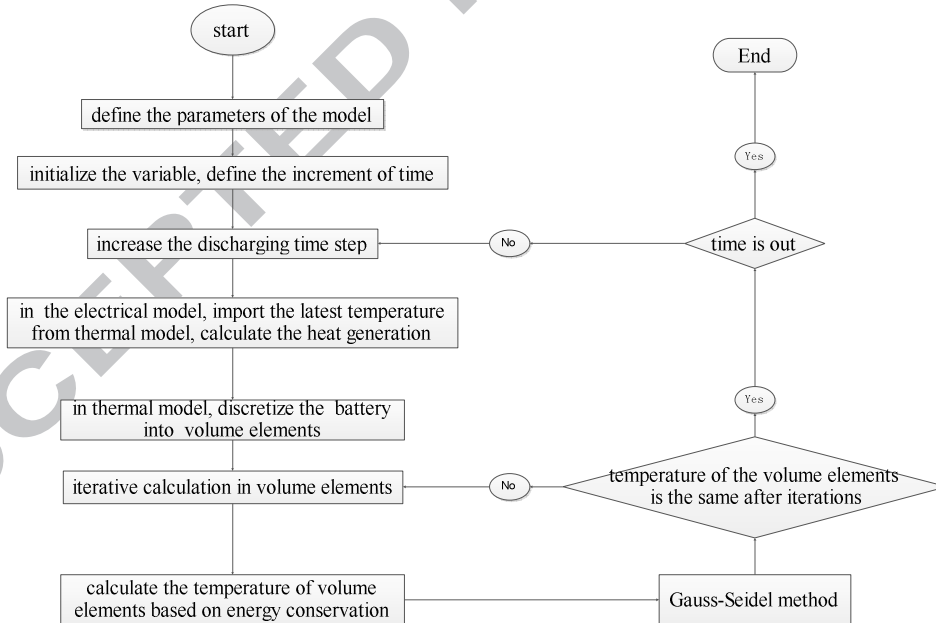


Figure 4 The flowchart of solution



## 3 Validation of model

### 3.1 Experimental validation of model

A commercial pouch battery (capacity 20Ah, nominal voltage 3.2V) was experimentally tested. The resistance was measured with chronopotentiometry method and the result was illustrated in figure 3. The apparatus was described in reference [29] where the work was done by our team. The batteries are charged by LAND-CT2001B, which is a charge and discharge multi-channel testing and controlling system, and are discharged by a programmable electronic load ITECH-IT8516. The operating currents, voltages, and temperature are monitored by data acquisition modules Agilent 34970A, and then processed by a host computer with built in software for programming charge and discharge. The battery was discharge at constant-current until it reached the cut-off voltage 2.7V. In order to validate the temperature field of the battery during discharging process, eight K-type thermocouples were placed on the front surface of the battery.

Figure 5(a) shows excess temperature profile at the point # 3 obtained by experiment and simulation at various discharge rate (0.5 C, 1.0 C, 1.5 C, 2.0 C. Here, the discharge rate C is equal to current of discharge divided by nominal capacity). Simulation results agree well with experiment. The excess temperature at high discharge rate increased faster because the heat generation is higher. Compared to the excess temperature at point #1 and point #2 illustrated in figure 6(b), prediction for point #3 is more precise with this model. That is because temperature at point #1 and point #2 is more susceptible to the tabs. The temperature of tabs is sensitive to contact resistance and ambient environment. The small deviation between the experiment and the simulation is due to the reversible heat which is gotten from the reference [11] during the calculation. Though the change trend of reversible heat is similar, there exists small deviation in

different references [30]. The calculation result may improve if the reversible heat was obtained from experiment with the same type of battery. Other reason is that the convective heat transfer coefficient is not precisely estimated. We assume the convective heat transfer coefficient is around  $4\text{W/m}^2$  under natural convection. The result will be better if the experiment is conducted under adiabatic condition.

The figure 5(b) illustrates the voltages of the battery at 1C discharge rate in simulation and experiment. They agree well with each other. The deviation at the beginning is bigger, relatively. That may be because the actual polarization resistance of battery is small than expected. At the beginning of discharge, the concentration gradient of  $\text{Li}^+$  is small and changes fast relatively. That may bring error to the result as the chosen step of the measure is a little bigger.

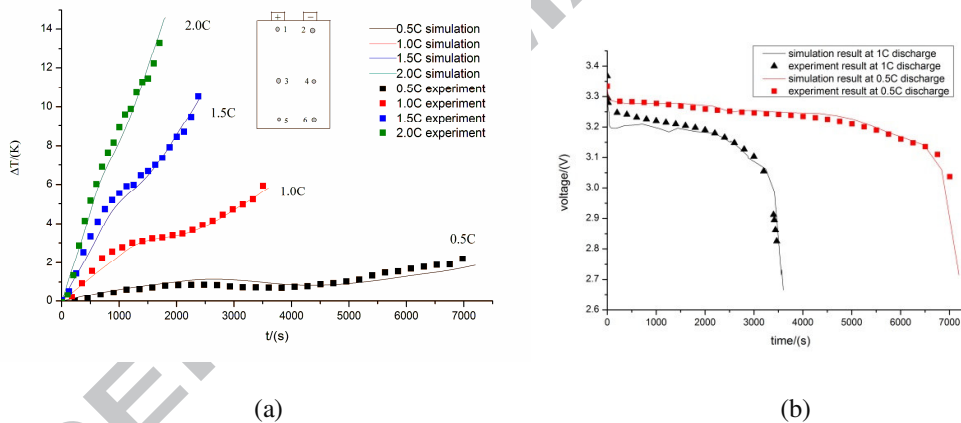


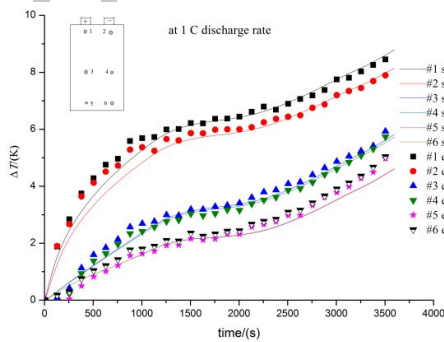
Figure 5 (a) comparison of simulated results of average surface temperatures at point #3 with experimental data during galvanostatic discharge (0.5 C, 1.0 C, 1.5 C, 2.0 C). (b) voltage curve at

1 C (0.5 C) discharge

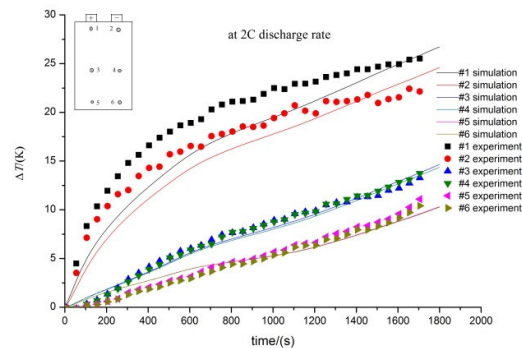
Temperature distribution was measured with K-type thermocouples at various rate of discharge. As illustrated in figure 6, 6 thermocouples were placed on the surface of the battery while two were placed on the tabs. Figure 6 shows the comparison of surface excess temperature

of different point during galvanostatic discharge (1 C, 2 C) under natural convection condition. An acceptable agreement of simulation and experimental curves can be seen. The deviation of simulation and experiment at point #1 and point #2 is bigger especially at 2 C discharge rate. This may be attributed to the influence of current collecting tabs. Temperature of positive tab is much higher than the battery. Convective condition and touch resistance can make a big difference to the temperature of tabs. Another reason is probably the effect of lateral current which is not rigorously considered in this model. The lateral current will flow to the tabs through the electrodes, not the current collectors[31].

The non-uniform of temperature distribution is quite serious especially in high discharging rate. At 2C discharge rate, the difference between the maximum and the minimum is as high as 14K, which would do great harm to the performance of the battery. Lifetime of battery will decrease seriously with so large temperature deviation. The temperature deviation will aggravate in battery modules because of uneven cooling condition. Therefore, necessary measurements should be taken to control not only the temperature but also the temperature non-uniformity of the battery.



(a)



(b)

Figure 6 Comparison of surface excess temperature of different point during galvanostatic discharge ( 1C on the picture a, 2C on the picture b) under natural convection condition. The simulated data is smooth while the experiment data is fluctuant

From the comparison of temperature curve, the slope at the middle stage (from SOC = 35% or  $t = 1260$  s to SOC = 70% or  $t = 2520$  s) is gentle at 1C discharge rate but it is steep at 2 C discharge rate. This is attributed to the influence of reversible heat which contributes more than 50% of the total heat generated (in Joules) at low discharge rate[32]. Reversible reaction heat is endothermic during discharging process and changes with SOC.

The model can also be used to calculate the temperature distribution and current density distribution during charge process. We just need to change the reversible heat as heating effect is different in the two processes (discharge and charge).

### 3.2 Comparison of two models

The model is convincing to predict the temperature field of the pouch battery when resistance is precisely measured and working condition is well described. The electro-thermal model is multilayer model as described before. Compared to the lumped model in which the thermal property of all layers is equal to the same equivalent value, the multilayer model well describes the inner structure of battery. Equivalent method for equivalent property is described in reference [28] in which the work was done by our team with the same type of battery. The simulation result of two models (lumped model and multilayer model) and the experiment result are shown in figure 7. According to the definition of irradiance uniformity in reference [33], temperature non-uniformity is defined as the ratio of deviation to the average temperature:

$$\phi = \frac{T_{cen,max} - T_{cen,min}}{T_{cen,max} + T_{cen,min}} \quad (12)$$

where the  $T_{cen,max}$  and  $T_{cen,min}$  are maximum and minimum centigrade temperature of battery.  $\Phi$  is equal to zero when the temperature of battery is uniform. From the comparison of the simulated result, temperature is more uniform in lumped model than in multilayer model. The reason of large deviation is that the heat conductivity of copper foil and aluminum foil is two orders larger in magnitude than that of electrode material. Large discrepancy between different layers will make the equivalent heat conductivity inaccurate to calculate the temperature field. Besides, calculation of equivalent heat conductivity and equivalent specific heat capacity is always independent from each other. As a result, equivalent thermal diffusivity ( $\lambda\rho c$ ) is overestimated in lumped model. The temperature non-uniformity  $\Phi$  of multilayer model is bigger than that of lumped model and shows more accurate. The analysis and conclusion can be extended to cylindrical battery because its inner physical structure is the same to pouch battery.

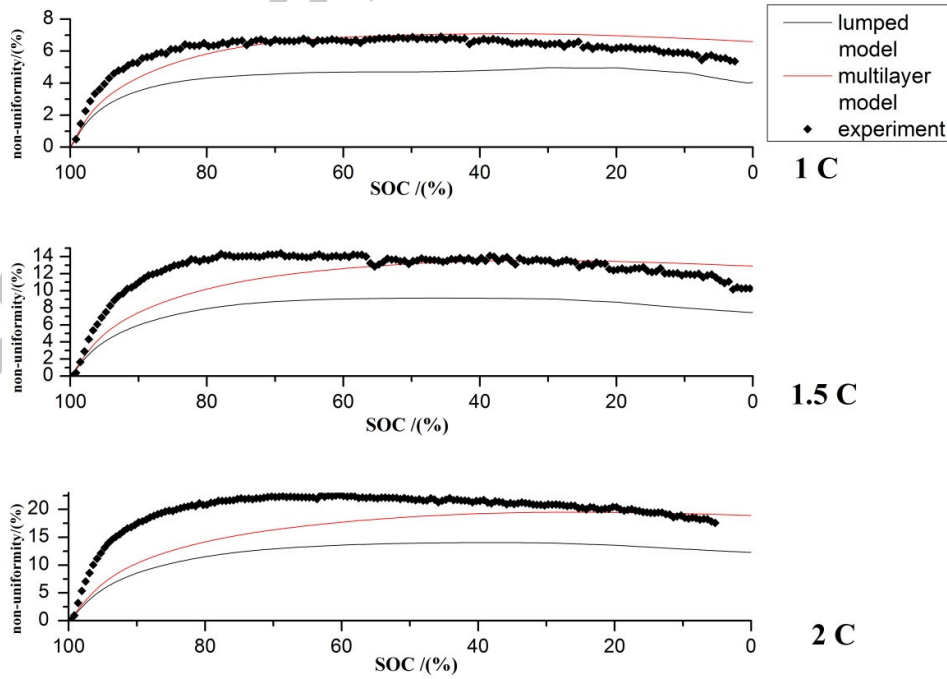


Figure 7 Comparison of temperature non-uniformity at various discharge rate (1 C, 1.5 C, 2 C)

## 4 Results and discussion

From the aforementioned description, the electro-thermal model is simple and the parameters are easy to measure. But the calculation result agrees with experiment.

### 4.1 Simulation result of temperature distribution

Numerical calculations were performed for the  $\text{LiFePO}_4/\text{graphite}$  pouch battery at 1 C, 2 C discharge rate to better understand the electrical and thermal characteristics of the battery. Figure 8 shows the excess temperature distribution in 3D cloud picture at 1 C discharge rate. The arrows indicate the direction and magnitude of the current flowing in the current collector layer of battery. Obviously the current flows into (out) the negative (positive) current collecting tab. The magnitude decreases with the increasing distance from the tab. That is because the local current flows across the electrodes of local cell element, then it will change the direction vertically and flows into the current collector[34]. All current from the local cell elements will gather in the tab. The current distribution is similar to the result of reference [9] and the model of reference [32].

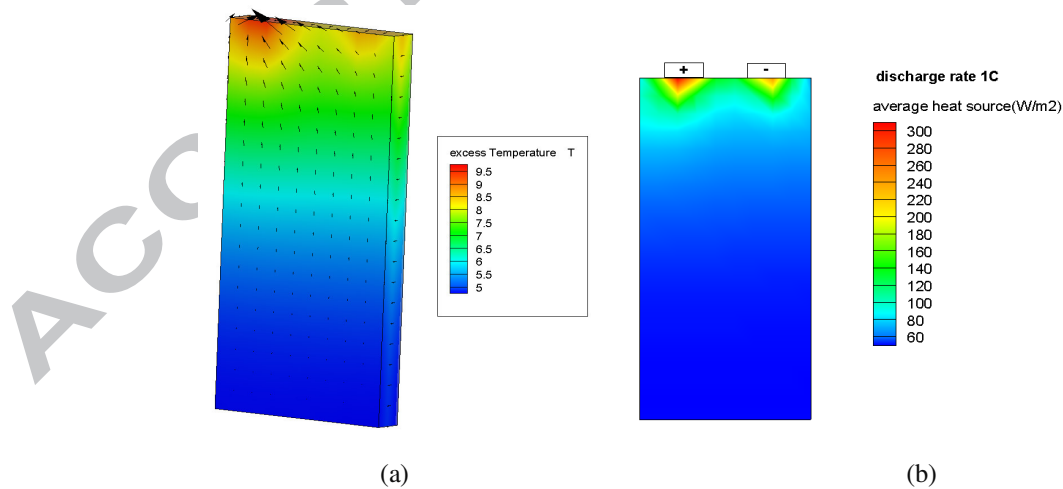


Figure 8 The excess temperature distribution under 1 C discharge at the end of discharge ( $t = 3600\text{s}$ ). The arrows indicate the direction and magnitude of the current. Tabs are not shown in the

picture. The ratio of x, y and z is adjusted (a). spatial distribution of average heat generation at 1C

discharge rate (b)

From the excess temperature distribution in figure 8, the excess temperature near the tab is higher. The temperature near the positive current collecting tab (point #1 as illustrated in figure 5) is higher than the temperature near the negative tab (point #2). It is because the electrical conductivity of the aluminum foil is lower than that of the copper foil. Extra simulation result have proved that the temperature deviation of point #1 and point #2 was less than 0.1K at 1C discharge rate when the current collectors were the same material (copper foil or aluminum foil).

It is interesting to notice the non-uniform of temperature distribution. As illustrated in figure 8, temperature increases gradually from the bottom to the top of battery. The maximum excess temperature is 8.7 K while the minimum is 4.6 K. The main reason is that the resistance of collectors and uneven current distribution will lead to non-uniform distribution of local voltage ( $U_{local}$ ) of cell elements, which will lead to non-uniform of overpotential distribution. Figure 9 shows the overpotential of local cell elements at the beginning of 1 C discharging. Generally, a greater overpotential will drive the electrochemical reaction of the electrode at a faster rate, which will lead to more electrochemical reaction heat[9]. The distribution of average heat generation at 1C discharge rate in figure 8(b) is quite similar to the distribution of overpotential in figure 9(a). The overpotential near the tabs is larger. The distribution is similar to temperature distribution.

The overpotential of local cell element is determined by equation (13):

$$\eta_{local} = U_{eq,local} - U_{local} \quad (13)$$

The open circuit potential of the local cell element  $U_{eq,local}$  depends on temperature, SOC and material of electrode/ electrolyte. At the beginning of discharging, the open circuit potentials of

the local cell elements are equal. The working voltage of local cell element  $U_{\text{local}}$  is determined by the electrical 2D network model built in section 2.2. Both the variation trend and the magnitude of overpotential in figure 8 is similar to that in the reference[9] .

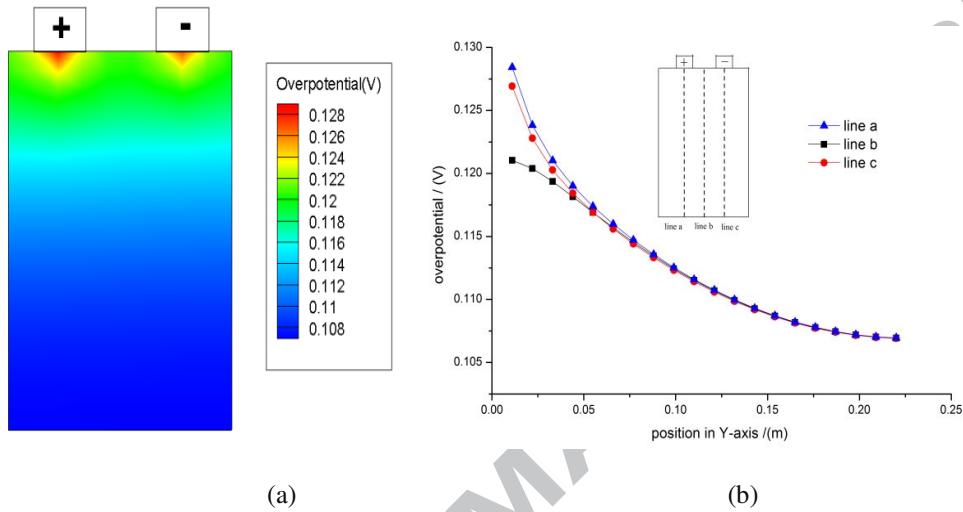


Figure 9 The overpotential distribution of local cell elements at the beginning of 1 C discharging. The distribution on the x-y plane (a) and the distribution along the a line (b)

## 4.2 Internal short circuit

The multilayer electro-thermal model can also be used to simulate the thermal behaviour of ISC before the battery temperature reaches 150°C. We just study the situation where the battery is under the condition of open circuit. Firstly, ISC at different state of charge (SOC) was simulated and we found that temperature of battery with higher SOC was higher, relatively. That was similar to the result of literature [35].



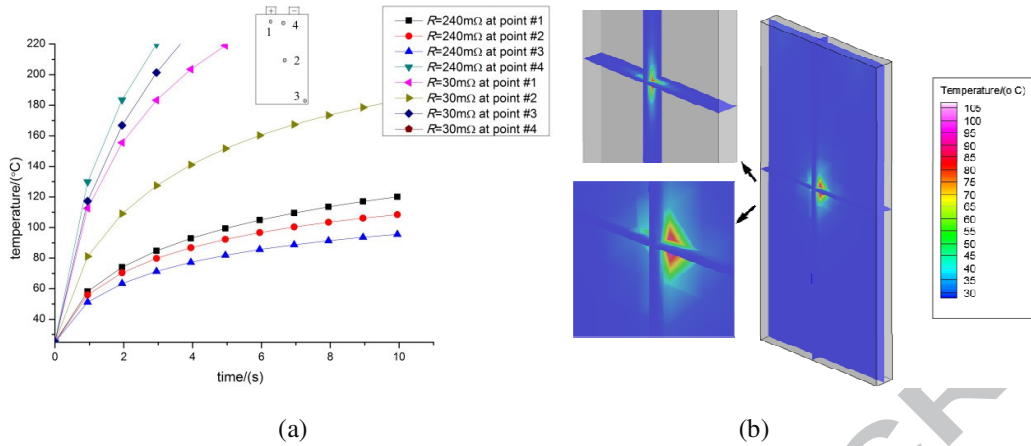


Figure 10 (a) Temperature of short-circuit cell element (anode-cathode short or anode-aluminum short) at different position. (b) Temperature distribution (t = 10 s) of battery suffering anode-cathode short at the middle position of the middle cell unit layer

Internal short circuit in a single cell unit is studied. We assume that one cell element in the middle cell unit (the 16<sup>th</sup> integrated layer and the adjoining collector layers) is short. Temperature of short-circuit cell element (anode-cathode short or anode-aluminum short) at different position is calculated and shown in figure 10(a). We assume the resistance ( $R_{\text{short}}$ ) of the an-ca short-circuit cell element is 240 m $\Omega$  while that of the an-Al short-circuit cell element is 30 m $\Omega$  [36]. The change trend is similar to that in the literature [36]. It can be seen that the an-Al short has a higher risk of thermal runaway when only a single layer is short. The result shows the same trend as described in the literature [19]. An-ca short will not go to thermal runaway though its temperature is higher than that at normal discharge. Temperature of short-circuit cell element at different position is different as illustrated figure 10(a). When the ISC cell element is near the tabs (#1), the spot temperature is the highest because of the influence of the collector resistance and the other cell units (different layers). Taking all cell elements (both from the short cell unit and the normal cell units) into consideration, the network circuit can be regarded as simple series circuit

connected with one short cell element, one equivalent resistance and one power source. Based on Thevenin's theorem, the equivalent resistance of the external circuit (not including the short cell element) can be calculated through the network model built in section 2.2. It is found that the equivalent resistance is the lowest when the #1 cell element is short. That means the voltage loss is the lowest in this way, which would lead to highest short current. Extra simulation also showed that the temperature of the (#4) short cell element at the middle of two tabs is the highest when the internal resistance of cell units becomes larger enough. That is because the equivalent resistance will become the lowest at (#4) short cell element if the internal resistance of cell units increases. From figure 10(a), temperature difference of an-Al short-circuit cell element at different position is larger than that of an-ca short-circuit cell element. That may be because magnitude of the short resistance of an-Al short is closer to that of collector resistance, relatively. As a result, an-Al short is more sensitive to the position. Another reason may attribute to the high current in an-Al short which can lead to high temperature.

Temperature distribution ( $t = 10$  s) of battery suffering anode-cathode short at the middle position of the middle layer is illustrated in figure 10. Temperature difference between the middle layer and the surface is as high as 55 K and that would make the detection of the ISC process difficult. The main reason is the low thermal conductivity of separator ( $k \approx 0.1 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ ) and electrode ( $k \approx 1 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ ) [9]. Through the simulation, the temperature of an-ca short and an-Al short element at middle layer/surface ( $t = 10$  s) is shown in table 2. The temperature of short element at the middle layer is the highest in the battery. It is found that increasing the thermal conductivity of integrated layer especially the separator layer can effectively relieve the heat spot effect of ISC. Moreover, increasing the thermal conductivity will make the detection of ISC easier.

Table 2 The temperature of an-ca short and an-Al short element at middle layer/surface ( $t = 10$  s)

thermal conductivity of integrated layer/( $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ )	0.45	1	10
Temperature of an-ca short element at the middle/surface /( $^{\circ}\text{C}$ )	72/26	56/29	37/34
Temperature of an-Al short element at the middle/surface /( $^{\circ}\text{C}$ )	150/28	99/36	60/51

Thermal behaviour of two ISC cell elements is simulated. When the two spots lie in the same cell unit, the heat generation increases but the highest temperature depends on the position of the two spots. The highest temperature will decrease if the distance of two spot is not close. That may be because the current is divided into two parts and the heat generation decreases at the spot. When the two spot lie in the same position of adjacent cell unit, the highest temperature would increase fast. Through simulation, the battery may go to thermal runaway if three adjacent cell units are an-ca short. Penetration experiment easily leads to fire because all cell units are short. The heat generation of nail penetration is much depending on the ISC resistance and contact resistance.

As we can see, the electro-thermal model can guide the safety design of battery. It would get high precision if the variation of ISC resistance is defined.

## 5 Conclusion

An electro-thermal coupled model for pouch battery is developed to study the thermal performance during normal discharge and internal short circuit process. The battery is discretized into many cell elements which are united as a 2D network equivalent circuit. The thermal model takes the multilayer structure into consideration.

The electro-thermal coupled model is validated with experiment and the simulation result can predict the temperature distribution of pouch battery. The temperature non-uniformity  $\Phi$  of

multilayer model is bigger than that of lumped model and shows more accurate. That is because equivalent thermal diffusivity ( $\lambda/\rho c$ ) may be overestimated when calculation of equivalent heat conductivity and equivalent specific heat capacity is independent from each other.

Current density distribution and excess temperature distribution are simulated with the model. The temperature non-uniformity is quantified and the reason of non-uniformity is analyzed. It is observed that non-uniformity of temperature distribution is large in the battery and it will aggregate with the discharge rate increasing. Non-uniform temperature distribution is attributed to the resistance of collectors and the uneven distribution of current density.

The electro-thermal coupled model is also used to study the performance of ISC and guide the safety design of battery. The temperature of the ISC element near tabs is the highest because the equivalent resistance of the external circuit (not including the ISC element) is the smallest when the resistance of cell units is small. It is also found that increasing the thermal conductivity of integrated layer can not only effectively relieve the spot effect of ISC but also make the detection of ISC easier.

## Abbreviation

$c$	heat capacity
$E_c$	cell potential for open-circuit
$F$	Faraday constant
$h_{\text{conv}}$	convective heat transfer coefficient
$h_{\text{rad}}$	radiation heat transfer coefficient
$I$	charge/discharge current
$I_k$	branch current flowing in or out of the node
$n$	the charge number pertaining to the reaction
$q$	total heat source
$q_j$	joule heat

$q_p$	irreversible electrochemical reaction heat
$q_r$	the reversible entropic heat
$R_{cp}$	concentration polarization resistance
$R_{ep}$	electrochemical polarization resistance
$R_{ohm}$	total ohmic resistances (including $R_o$ , $R_{ta,A}$ , $R_{ax,A}$ , $R_{ta,C}$ and $R_{ax,C}$ )
$R_p$	polarization resistance
$R_{short}$	resistance of internal short circuit
$t$	time
$T$	temperature
$T_{am}$	ambient temperature
$T_{cen,max}$	maximum centigrade temperature of battery
$T_{cen,min}$	minimum centigrade temperature of battery
$V_k$	the voltage of the electric element
$U_{eq,local}$	open circuit potential of the local cell element
$U_{local}$	working voltage of local cell element
$\tau_{ep}$	time constant
$\tau_{cp}$	time constant
$\rho$	density
$\lambda$	heat conductivity
$\varepsilon$	surface emissivity
$\sigma$	Stefan-Boltzmann constant
$\Phi$	temperature non-uniformity
$\eta_{local}$	overpotential of local cell element
$\Delta H$	reaction enthalpy change
$\Delta G$	electrical work

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ACCEPTED MANUSCRIPT

Table caption

Table 1 thermal property of different layers of battery

Table 2 The temperature of an-ca short and an-Al short element at middle layer/surface ( $t = 10$  s)

ACCEPTED MANUSCRIPT

## Figure caption

Figure 1 Layer structure of pouch battery (the ratio of size is adjusted for better illustration)

Figure 2 Illustration of the cell element equivalent circuit (a) and 2D network equivalent circuit (b)

Figure 3 The potential response of battery (a) and resistance of battery (b) measured with chronopotentiometry method

Figure 4 the flowchart of solution

Figure 5 (a) comparison of simulated results of average surface temperatures at point #3 with experimental data during galvanostatic discharge (0.5 C, 1.0 C, 1.5 C, 2.0 C). (b) voltage curve at 1 C (0.5 C) discharge

Figure 6 Comparison of surface excess temperature of different point during galvanostatic discharge (1C on the picture a, 2C on the picture b) under natural convection condition. The simulated data is smooth while the experiment data is fluctuant

Figure 7 Comparison of temperature non-uniformity at various discharge rate (1 C, 1.5 C, 2 C)

Figure 8 The excess temperature distribution under 1 C discharge at the end of discharge ( $t = 3600s$ ). The arrows indicate the direction and magnitude of the current. Tabs are not shown in the

picture. The ratio of x, y and z is adjusted (a). spatial distribution of average heat generation at 1C discharge rate (b)

Figure 9 The overpotential distribution of local cell elements at the beginning of 1 C discharging.

The distribution on the x-y plane (a) and the distribution along the a line (b)

Figure 10 (a) Temperature of short-circuit cell element (anode-cathode short or anode-aluminum short) at different position. (b) Temperature distribution ( $t = 10$  s) of battery suffering anode-cathode short at the middle position of the middle cell unit layer

Highlight:

2D network equivalent circuit considers the interplay of cell units.

The temperature non-uniformity  $\Phi$  of multilayer model is bigger than that of lumped model

The temperature non-uniformity is quantified and the reason of non-uniformity is analyzed.

Increasing the thermal conductivity of the separator can effectively relieve the heat spot effect of

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