A Probe into the Accuracy of Thermal Runaway Simulation Model of Lithium-ion Battery under Adiabatic Condition

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Abstract. For the safety problem of lithium-ion battery, thermal abuse early warning is needed to be established, for which the main point is to analyze the mechanism and to establish the thermal abuse model of single lithium-ion battery. This article used 10Ah ternary lithium-ion battery as the research object and used simulation software to build thermal abuse simulation model. The accuracy of simulation model was verified by comparing the temperature simulation result with temperature data from adiabatic environment thermal abuse experiment. According to the results, heating trend of temperature of the simulation under adiabatic condition is approximately the same as that of ARC adiabatic test temperature. Before the end stage of self-heating period, the error remains within 5%; at the end of self-heating stage, the error is around 5-10%; after entering thermal runaway stage, the error can reach 36%. Missing the equation of reactions between electrode and additive, and internal short-circuit is the main reason causing the error of the simulation result.

1. Introduction
The lithium-ion battery is widely used in such fields as portable equipment, electric vehicles, power grid energy storage, etc., and plays an irreplaceable role[1]. According to incomplete statistics, there were 29 fire accidents have occurred in South Korea and 3 fire accidents at energy storage power stations have been reported in China by the end of 2020. Huge property damage and casualties have been caused by these accidents, so the importance of safety research on lithium-ion battery increases.

In the Paper, the 10Ah ternary lithium-ion battery monomer will be taken as the research object, the temperature change in the thermal runaway of the battery is analyzed by using the accelerating rate calorimeter (ARC), and at the same time, the thermal runaway process of the battery will be simulated by using the battery thermal runaway model of simulation software. With the two experimental results being compared, the simulation accuracy of the software model will be analyzed.

2. Establishment of model

2.1. Characteristic parameters of the battery and material
The establishment of this battery model is based on the fact that 10Ah ternary battery produced by a company is taken as the research object. The main parameters of the battery sample are shown in Table 1.
Table 1 Main Parameters of Lithium-ion Battery Sample

| Name                  | Parameter                                      |
|-----------------------|-----------------------------------------------|
| Anode material system | Li(Ni_{1/3}Co_{1/3}Mn_{1/3})O_2               |
| Cathode material system | Graphite                                      |
| Outline dimension (mm) | 88*56*25                                      |
| Mass (kg)             | 0.25Kg                                        |
| Mean density (kg/m³)  | 2780                                          |
| Mean specific heat capacity (J/(kg*K)) | 1099                                           |
| Heat conductivity coefficient (W/(m*K)) | $\lambda$ = $\lambda_x$ = 3.957, $\lambda_y$ = 0.887 |

2.2. Experiment of thermal runaway of the battery
In the Paper, the adiabatic thermal runaway experiment of the battery is carried out by ARC(BTS-500).

2.3. Thermal runaway simulation of battery monomer
The model is meshed with a hexahedral structure, and the grid model as shown in Fig. 1 is established. Among them, a temperature monitoring point is set at the surface center of the battery body and the maximum external contact surface.

Fig.1 Thermal Runaway Model of Monomer

The default thermal runaway model in the simulation software is selected, and its model parameters are shown in Table 2:

Table 2 Thermal Runaway Model Parameters

|        | A (1/s) | E (J/mol) | m | n | H (J/g) | W (g/m³) | α   | c   |
|--------|---------|-----------|---|---|---------|----------|-----|-----|
| SEI    | 166700  | 135080    | 1 | - | 257     | 610400   | 0.15| -   |
| Cathode| 2500    | 135080    | 1 | - | 1714    | 610400   | 0.75| -   |
| Anode  | 6667    | 139600    | 1 | 1 | 314     | 1221000  | 1   | 0.04|
| Electrolyte | 5.14×10^{15} | 274000   | 1 | - | 155     | 406900   | 1   | -   |

2.4. Characteristic parameters of the battery
Control equation of the calculation model of thermal runaway of batteries
In essence, the thermal runaway of the lithium-ion battery is the self-heating reaction inside the active material under high temperature, which leads to the rapid temperature rise and the release of a large amount of heat. Complex unsteady heat transfer processes will occur between the process and the outside world. According to the law of conservation of energy, Equation (1) is met in the process:

$$\rho C_p \frac{\partial T}{\partial t} = \lambda_x \frac{\partial^2 T}{\partial x^2} + \lambda_y \frac{\partial^2 T}{\partial y^2} + \lambda_z \frac{\partial^2 T}{\partial z^2} + Q(t)$$  (1)

Among them, $\rho$ represents the unit density of the battery, $C_p$ represents the unit heat of the battery, $\lambda_k$ represents the thermal conductivity of the unit, the subscript $k$ represents direction, $T$ represents temperature, $t$ represents time, and $Q(t)$, being the heat component in the equation, represents the sum of the heat generation rate and the heat loss rate. With the fact that the thermal runaway reaction laboratory for the battery is conducted in an adiabatic environment being taken into consideration, $Q(t)$
only represents the heat generation rate in the thermal runaway process. The heat release in the thermal runaway process of the battery is mainly divided into four steps, where \( Q(t) \) can be regarded as the sum of the four steps above:

\[
Q(t) = Q_{\text{SEI}}(t) + Q_{\text{neg}}(t) + Q_{\text{ele}}(t) + Q_{\text{pos}}(t)
\]

(2)

Here, \( Q_x(t) \) represents the reaction heat production of component \( x \), the subscript \( x \) may represent SEI, cathode, electrolyte, and anode [2-3].

3. Results and discussion

3.1. Adiabatic thermal runaway experiment results

From Fig. 2, which shows the experimental results of adiabatic thermal runaway carried out on the samples by using ARC, it can be seen that there are totally 4 stages from the beginning of heating to the end of cooling of thermal runaway:

1) Heating stage: At 25-82.8°C, this stage occurs. Adiabatic heating of the lithium-ion battery.
2) Self-heating stage: This process occurs at a temperature range of 82.8-147.8°C and lasted for about 17,300s. As a reaction has taken place inside the battery, the temperature of the battery begins to rise slowly.
3) Thermal runaway stage: At a temperature range of 147.8-458°C, this stage occurs and lasts for about 490s. Because the reactions inside the battery accumulate, the internal structure of the battery is completely destroyed, resulting in an internal short circuit and the release of a large amount of heat.
4) Cooling stage: ARC stops providing the adiabatic environment, so the temperature at the monitoring point drops significantly after the runaway heat of the battery runs out.

![Fig. 2 ARC Experimental Results](image)

3.2. Simulation results of thermal runaway of battery monomer

In Fig. 3, the relationship between temperature and time in the process of thermal runaway obtained by simulation software is shown. In the figure, it can be clearly seen that after the temperature enters the self-accelerating stage at 80.15°C for 17,350s, the temperature rising rate reaches 2°C/min, then it enters the thermal runaway stage. At this time, the temperature is 137°C. The thermal runaway finally reaches the maximum temperature after 480s, and the reaction of the active materials inside the battery is complete. This shows that the established thermal runaway model is basically consistent with the temperature, duration time of self-heating, and thermal runaway when the ARC experiment enters the self-heating stage.
3.3. Verification and error analysis of simulation results

To carry out further analysis of the difference between the simulation model and the experimental results, the error analysis of the temperature data obtained by the two methods is shown in Fig. 4. Among them, the model error is calculated according to Equation (3)[4]:

\[
\text{Error} = \left( \frac{T_{ARC}(t_i) - T_{sim}(t_i)}{T_{ARC}(t_i)} \right) \times 100\%
\]

Among them, Error represents the error of the mode, \( T_{ARC}(t_i) \) represents the temperature value of ARC experiment at \( t_i \) time, \( T_{sim}(t_i) \) represents the simulation calculation analog value at \( t_i \) time. From the figure, it can be seen that there is a certain error in the initial stage of self-heating, which is caused by the difference in temperature between the simulation model and the battery ARC tested battery when it enters the self-heating stage. When both of them enter the self-heating reaction, the error remains within 5% as of 15,700s, indicating that the simulation results are basically consistent with the test results in the self-heating stage of the battery. However, after 15,700s, the temperature of the battery at this time is close to that of battery thermal runaway, and the error at this time increases significantly, with the maximum error being about 36%, which is caused by the fact that the control equation in the software does not include the heat generated by melting the membrane and releasing heat from a short circuit in the battery, so there will be a certain of error when it is close to thermal runaway.

On the whole, the software simulation results are basically consistent with the ARC experimental results, and the established model is much reliable. However, it is still necessary to further improve the thermal runaway model, and more factors affecting thermal runaway should be taken into consideration, so as to further improve the accuracy of the simulation.
4. Conclusions
In the Paper, the accuracy of the simulation results of battery thermal runaway is studied. With the simulation results being compared with the ARC adiabatic test results, the following conclusions can be drawn:

(1) The heating trend of temperature of the simulation under adiabatic condition is approximately the same as that of ARC adiabatic test temperature. After 5 hours of self-heating period, the battery enters thermal runaway stage and all energy of the battery would be released after 10 minutes.

(2) Before the end stage of self-heating period, the error remains within 5%; at the end of self-heating stage, the error is around 5-10%; after entering thermal runaway stage, the error can reach 36%.

(3) From the result, missing the equation of reactions between electrode and additive, and internal short-circuit is the main reason causing the error of the simulation result. It is necessary for researchers to analyze the electrical energy release efficiency and chemical reactions during the thermal runaway process, add more relative equations into the software to further improve the accuracy of the results.

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