ABSTRACT: To solve the problem of thermal runaway is one of the necessary conditions for the commercialization of lithium-ion batteries. In order to further explore the reaction mechanism of thermal runaway of lithium-ion batteries, a thermal model is built by using a variety of side reactions to further study the inhibition of temperature on thermal runaway. The results show that thermal runaway is triggered by the heat generation of negative material reaction when it is heated to 473.15 K; lower heat dissipation temperature (273.15 K) cannot effectively inhibit the occurrence of thermal runaway.

1. INTRODUCTION
Lithium-ion batteries have a wide range of applications in vehicles, homes, and portable mobile devices and provide solutions to reduce environmental pollution. However, thermal runaway of a lithium-ion battery is still one of the difficulties to be overcome in its large-scale application. For lithium-ion batteries to be fully commercialized, research on thermal runaway is necessary. A variety of conditions can lead to thermal runaway, including short circuits, extrusion, heat abuse, and so on. For example, external environmental factors and failure of some automotive systems can lead to heat abuse. In order to reveal the mechanism of thermal runaway, researchers had developed experiments including high-speed imaging, battery piercing, and simulation model with various dimensions. This article explains the thermal runaway process of the battery through the side reactions of different components of the battery.

Abraham et al. used temperature as a marker to distinguish the side reactions of anode, cathode, and electrolyte in the process of thermal runaway of lithium-ion batteries, among which the characteristic temperatures were 90, 140, and 180 °C, but the article does not focus on electrothermal phenomena. There are still many points worth discussing about the triggering factors and the changing process of thermal runaway. The following studies are mainly carried out with short circuit as the triggering factor or the main factor. Feng et al. segmented the thermal runaway process according to the composition of the battery, which contained a large amount of heat generated by an internal short circuit. Ping et al. studied the thermal runaway process of lithium cobalt-oxide batteries. With the decomposition of the separator and the SEI (solid electrolyte interface) film, the battery short-circuit and the side reactions in anode, cathode, and electrolyte caused the temperature to rise rapidly. Zheng et al. believed that it is the internal short circuit that causes the decomposition of materials such as anode and cathode, resulting in side reactions that generate a large amount of reaction heat.

Therefore, it is worth trying to study the thermal runaway process of lithium-ion batteries by using the idea of segmentation. The adjustment of the battery's thermal management system has limits. When the thermal management system can control the temperature below 60 °C, some exothermic side reactions do not occur. If the battery temperature cannot be successfully controlled below 60 °C, the probability of thermal runaway will be greatly increased, which means there will be qualitative change of the battery heat release. Too many studies focus on two points: improving the thermal management system to make the temperature not get out of the control area and the mechanism to make the temperature rise sharply during thermal runaway. However, a few people have focused on the inhibition of lithium-ion battery thermal runaway. In the early stage of thermal runaway, not all substances begin to release heat, so the temperature...
does not rise sharply. This article explores the possibility of suppressing thermal runaway by temperature and hopes that low temperature can stop the thermal runaway process.

In this paper, COMSOL was used to build a model of the lithium-ion battery, and different dissipation temperatures were used to suppress the thermal runaway process, and the process of internal side reactions of the SEI film, anode, cathode, and electrolyte were observed. Meanwhile, thermal runaway is triggered by the heated local area of the battery surface because the battery is usually locally heated to thermal runaway by the adjacent battery which is in thermal runaway.

2. RESULTS AND DISCUSSION

2.1. Thermal Runaway Process with No Inhibition. In this section, the battery is heated on one-third surface area of the positive current collector to 473.15 K to study the characteristic of the whole thermal runaway process. The temperature rise curve of the lithium-ion battery is shown in Figure 1. It can be seen that there are two turning points in the curve and one temperature peak. There was a sudden change in temperature at about 200 and 300 s. The temperature peak appears at approximately 350 s, which reaches 550 K, and then the temperature decreases gradually until 475 K. The temperature does not get back to the ambient temperature because the simulation time stops at 1000 s, and it will decrease again if the simulation continues. From the whole curve, it can be seen that thermal runaway is over in only 350 s, and the temperature rises very sharply.

Figure 2 shows the heat production curve of each side reaction during the whole thermal runaway process. It can be seen from the picture that all side reactions happen and all the reactants are finished. The heat generated by the reaction of the positive material is much higher than that of the other three reactions, which is about 3 times that of the reaction of the negative material. The SEI decomposition reaction (SEI) is the earliest reaction, starting at about 200 s, and the reactions of the positive material (Pos) and the negative material (Neg) and the electrolyte reaction (Ele) basically take place at the same time, around 300 s. The latter three reactions almost end at the same time, around 350 s.

According to the results in Figures 1 and 2, the main heat source of the temperature rise when the battery thermal runaway happens is the reaction of the positive electrolyte material and the reaction of the negative electrolyte material. The main reason for the first temperature rise is external high-temperature heating; the first temperature inflection point is caused by the SEI film decomposition reaction, and the second temperature inflection point is caused by the positive material reaction and the negative material reaction and the electrolyte reaction. The cloud image in Figure 3 shows the situation near the end of the thermal runaway process, at about 400 s. The temperature of the upper part of the battery is higher, and the temperature of the lower part is lower. The overall temperature at the heating position is basically the same, and the remaining parts gradually decrease around the heating position at the bottom and form low-temperature areas on both sides.

In this paper, the thermal runaway process of a lithium-ion battery is segmented according to the temperature turning point and the temperature peak in the above figure:

Stage 1: Stage one starts at 293.15 K. At this point, the temperature rise of the battery is mainly caused by external heating, and the temperature change caused by the heat of reaction is very small.

Stage 2: Stage two starts from 450 K. At this time, as the temperature difference between the battery and the ambient temperature increases, the heat transfer between the battery and the surrounding environment increases, so the temperature rise slows down. The side reactions of SEI film also begin to exotherm at this stage.

Stage 3: Stage three starts from 475 K. At this stage, the SEI film decomposition reaction is over, and the reactions of
positive material and negative material and electrolyte reaction take place. These three reactions generate much more heat than the SEI film decomposition reaction which make the temperature rise sharply.

Stage 4: Stage four starts from 550 K and ends until the temperature is equal to the ambient temperature. At this stage, all reactions are over, and the battery is only in the heat dissipation process.

2.2. Inhibition with 293.15 K at 180 s. As can be seen from the above section, thermal runaway starts from SEI decomposition reaction (200 s). Based on these results, this section studies the inhibition of temperature rise caused by runaway heat. The cooling temperature is 293.15 K, and the position is part of the bottom surface of the negative material. The cooling starts at 180 s, a little earlier than 200 s. Figure 4 shows the change curve of battery temperature under this inhibition mode. As can be seen from the figure, the temperature curve of the battery shows obvious changes. The first temperature inflection point disappears, and the battery directly enters the fourth stage after reaching 450 K, and an insignificant temperature peak is formed at 450 K. This shows that this kind of inhibition mode has obvious effect, and the temperature rise of the battery is obviously reduced.

Figure 5 shows the heat production curve of each side reaction under this inhibition mode. By comparing with the temperature changes during the previous thermal runaway, it can be found that the main reaction occurred between the SEI film and the negative electrode. The negative material is not finished because it only generates half the heat of the SEI film decomposition reaction. The heat generated by the reaction of the positive material is much higher than that of the other three reactions, which is about 3 times the reaction of the negative electrolyte material. The SEI decomposition reaction is the earliest reaction, starting at about 200 s, and the reactions of positive material and negative material and electrolyte reaction basically take place at the same time, around 300 s. The latter three reactions almost end at the same time, around 350 s.

By observing Figures 4–6, thermal runaway can be suppressed obviously by cooling the bottom surface, and the thermal runaway of the battery is segmented:

Stage 1: Stage one starts from 293.15 K. At this point, the temperature rise of the battery is mainly caused by external heating, and the temperature change caused by the heat of reaction is very small.

Stage 2: Stage two starts from 450 K. At this stage, the side reactions are winding down, and the temperature decreases gradually.

Stage 3: Stage three starts from 440 K. At this stage, the temperature remains stable because of the balance between heat dissipation and heat generation.

2.3. Inhibition with 283.15 K at 180 s. As can be seen from the above section, heat dissipation before the starting point of SEI membrane decomposition reaction can effectively inhibit the thermal runaway of lithium-ion batteries. In this section, the heat dissipation temperature is further decreased to 283.15 K when other conditions remain unchanged. Figure 7 shows the change curve of battery temperature under this
inhibition mode. By observing the figure, when the heat dissipation temperature is further decreased to 283.15 K, the temperature curve of the battery almost shows no change. The temperature of the battery still rises at an inflection point (a little earlier before 200 s).

Figure 8 shows the heat generation curves of each reaction under this heat dissipation mode. It can be seen from the figure that the internal heat production reaction of the battery is SEI membrane decomposition reaction and negative electrode material reaction as well, and the heat production of SEI decomposition reaction is 3 times that of negative electrode heat production. The curves of the reactions are similar to those in the previous section, which means 10 °C lower, and almost shows no effect on the reaction.

Figure 9 shows the battery temperature distribution of the analysis under this inhibition mode. As can be seen from the figure, the temperature distribution of the battery at the end of the analysis is similar to that in the previous section, except that the highest temperature is slightly lower and the battery status is the same as the previous one too.

Stage 1: Stage one starts from 293.15 K. As in the previous case, the heat source at this stage is mainly caused by external heating, causing the battery temperature to rise.

Stage 2: Stage two starts from 450 K. At this stage, the side reactions are winding down and the temperature decreases gradually.

Stage 3: Stage three starts from 440 K. At this stage, the thermal inhibition produced an effect, some exothermic side reactions did not occur, and thermal equilibrium is reached.

2.4. Inhibition with 273.15 K at 180 s. As can be seen from the previous section, 10 °C lower heat dissipation temperature failed to prevent the heat from getting out of control. In this section, the heat dissipation temperature is further decreased to 273.15 K for analysis, and the remaining conditions remain unchanged. Figure 10 shows the change curve of battery temperature under this inhibition mode. It can be seen from the figure that when the heat dissipation temperature is further decreased to 273.15 K, the temperature curve of the battery changes. The temperature rises in the same trend, but it reaches the peak value at about 180 s, which means that further lower heat dissipation temperature has an effect on inhibition.

Figure 11 shows the heat generation curves of each reaction under this heat dissipation mode. It can be seen from the figure that the internal heat generation reaction of the battery is SEI decomposition reaction and negative electrode material reaction, and it is similar to the result in the previous section that the heat production of SEI decomposition reaction is 3 times that of negative electrode heat production. The starting time of the two reactions are almost the same, which further shows that the inhibition has not improved a lot.

Figure 12 shows the battery temperature distribution of the analysis under this inhibition mode. As can be seen from the figure, the temperature distribution of the battery at the end of the analysis is similar to that in the previous section, except that the highest temperature is slightly lower and the battery status is the same as the previous one too.
Figure 12. Temperature distribution at the end of inhibition.

figure, the temperature distribution of the battery at the end of the analysis is similar to that in the previous section. As a result, the battery status is divided into the same three stages. Comparing the simulations with no suppression and different temperature suppressions, it can be found that temperature suppression will have a significant effect on the thermal runaway process, reduce the temperature exotherm peak, and change the turning point, but not enough to prevent thermal runaway. The difference between the simulation results of different temperatures is not large, which shows that the single temperature reduction has basically no effect.

3. CONCLUSIONS

In this paper, by adding different side reactions to the negative electrolyte, positive electrolyte, separator, and SEI film of the lithium-ion battery, the thermal runaway process of lithium-ion batteries was studied, and the mechanism revealed the suppression of thermal runaway of the batteries by changing the temperature. Through the simulation, the curves of the heat generation change of the reaction of each part of the battery and temperature evolution were obtained. From the simulation results, the following conclusions can be drawn: external heating can cause thermal runaway and the triggering of the thermal runaway process is mainly caused by the side reaction of the anode material; thermal inhibition could change the time of heat generation and the peak value of heat release in the process of thermal runaway, which was related to the trigger temperature of the reaction; at the same time, the evolution curve of the battery temperature with time also had some change; the heat generation of the positive pole was significantly reduced, whereas the heat release of the negative pole and SEI film was more obvious; lower heat dissipation temperature (273.15 K) cannot effectively inhibit the occurrence of thermal runaway.

4. RESEARCH METHODOLOGY

4.1. Thermal Abuse Reaction Model. In this paper, a thermal model is established based on the side reactions of the lithium-ion battery during thermal runaway. The side reactions are SEI membrane decomposition reaction, anode reaction, cathode reaction, and electrolyte decomposition reaction to describe the change process of heat abuse over time. In the literature, heat abuse was also studied through various side reactions.20−27

| symbol | description | value |
|--------|-------------|-------|
| A_{SEI} | SEI film decomposition frequency factor | 1.667 \times 10^{15} \text{s}^{-1} |
| A_a | anode-electrolyte frequency factor | 2.5 \times 10^{10} \text{s}^{-1} |
| A_{a,1} | positive-electrolyte decomposition frequency factor | 1.75 \times 10^{10} \text{s}^{-1} |
| A_{a,2} | positive-electrolyte decomposition frequency factor 1 | 1.077 \times 10^{12} \text{s}^{-1} |
| A_e | electrolyte decomposition frequency factor 2 | 5.14 \times 10^{15} \text{s}^{-1} |
| E_{SEI} | SEI film decomposition activation energy | 1.3508 \times 10^7 \text{J mol}^{-1} |
| E_{a,e} | anode-electrolyte activation energy | 1.3508 \times 10^7 \text{J mol}^{-1} |
| E_{a,pe.1} | NCM decomposition activation energy one | 1.1495 \times 10^7 \text{J mol}^{-1} |
| E_{a,pe.2} | NCM decomposition activation energy two | 1.5888 \times 10^7 \text{J mol}^{-1} |
| E_{e,se} | electrolyte decomposition activation energy | 2.74 \times 10^7 \text{J mol}^{-1} |
| \epsilon_{SEI} | initial value of the dimensionless number of SEI | 0.15 |
| \epsilon_{eel} | initial value of negative electrolyte dimensionless number | 0.75 |
| \alpha_0 | initial value of percentage of the reaction | 0.04 |
| \epsilon_0 | initial value of dimensionless number related to concentration | 1 |
| m_{SEI} | reaction order | 1 |
| m_a | reaction order | 1 |
| m_{pe.1} | reaction order | 1 |
| m_{pe.2} | reaction order | 1 |
| m_e | reaction order | 1 |
| H_{SEI} | SEI film decomposition heat release | 257 \text{J g}^{-1} |
| H_a | anode-electrolyte heat release | 1714 \text{J g}^{-1} |
| H_{a,1} | NCM decomposition heat release one | 77 \text{J g}^{-1} |
| H_{a,2} | NCM decomposition heat release two | 84 \text{J g}^{-1} |
| H_e | electrolyte decomposition heat release | 155 \text{J g}^{-1} |
| W_c | specific carbon content | 6.104 \times 10^5 \text{gm}^{-3} |
| W_p | specific NCM content | 1.221 \times 10^5 \text{gm}^{-3} |
| W_e | specific electrolyte content | 4.069 \times 10^5 \text{gm}^{-3} |

4.1.1. SEI Film Decomposition Reaction. After the manufacture of lithium-ion batteries is completed, the negative electrolyte is coated with an SEI film that isolates the cathode material from the electrolyte. This membrane will undergo decomposition reaction when the temperature reaches 353.15−393.15 K, and the heat of reaction will be released at the same time. The reaction formula is as follows:20

\[ k_{SEI}(T, \epsilon_{SEI}) = A_{SEI} \exp\left(-\frac{E_{a,SEI}}{RT}\right) \epsilon_{SEI}^{m_{SEI}} \]  

Heat generation equation

\[ Q_{SEI} = H_{SEI} W_c k_{SEI} \]  

where \( k_{SEI} \) is the reaction kinetic parameter; \( T \) is the temperature; \( \epsilon_{SEI} \) is a dimensionless number related to the content of lithium ions; \( R \) is the gas constant; \( Q_{SEI} \) is the heat production rate. More detailed parameters can be found in Table 1.

4.1.2. Negative Electrolyte Reaction. With the decomposition reaction of the external SEI film, the negative electrode will generate the side reaction of heat production at about 393.15 K. The following eq 3 is the negative electrode reaction equation.25 Equation 2 also applies to the calculation of heat production in this part.
where $k_{\text{ne}}$ is the reaction kinetic parameter; $T$ is the temperature; $c_{\text{ne}}$ is a dimensionless number associated with the content of lithium ions in the negative electrode; $t_{\text{SEI}}$ is the dimensionless number of the SEI film thickness; $t_{\text{SEI,ref}}$ is the initial value of the dimensionless number of the SEI film thickness. More detailed parameters can be found in Table 1.

In this paper, it is considered that the negative electrode only occurs after the partial SEI film reaction, and the judgment value is $c_{\text{SEI}} < 0.1$.

### 4.1.3. Positive Electrolyte Reaction.

In this paper, it is considered that the temperature over 393.15 K is one of the conditions for side reactions in the positive electrode. The following equation is the positive electrode reaction equation.

$$k_{\text{pe}}(T, c_{\text{pe}}) = A_{\text{pe}} \exp\left(-\frac{E_{\text{a,pe}}}{RT}\right)$$  \hspace{1cm} (4)

where $k_{\text{pe}}$ is the reaction kinetic parameter; $T$ is the temperature; $c_{\text{pe}}$ is a dimensionless number related to the concentration; $\alpha$ is the percentage of the reaction. More detailed parameters can be found in Table 1. In the positive electrolyte runaway reaction, the exothermic process has two maximum values, which can be shown by this side reaction.\textsuperscript{26,27}

### 4.1.4. Electrolyte Decomposition Reaction.

The electrolyte generally produces major side reactions when the temperature is higher than 473.15 K. The following equation is the electrolyte decomposition reaction equation.

$$k_{\text{e}}(T, c_{\text{e}}) = A_{\text{e}} \exp\left(-\frac{E_{\text{a,e}}}{RT}\right)$$  \hspace{1cm} (5)

where $k_{\text{e}}$ is the reaction kinetic parameter; $T$ is the temperature; $c_{\text{e}}$ is a dimensionless number related to the concentration. More detailed parameters can be found in Table 1.\textsuperscript{24}

### 4.2. Geometric Model.

In order to reduce the computational cost of the model, the real battery structure is partially simplified. The simplified model was built by COMSOL Multiphysics. The model is simplified by integrating the various thin layers, including the current collector, the positive electrolyte, the negative electrolyte, the separator, and the electrolyte as well as the battery tabs. There will be different side reactions in different regions, and the heat from the reaction will be conducted between these regions. Figure 13 shows the geometric model used in this article.

### 4.3. Boundary Condition.

The cooling location is part of the bottom area of the battery, and three cooling temperatures and beginning times are considered. The specific values can be found in Table 2. The battery conducts heat with the surrounding air. The heating position and temperature measurement point of the battery are shown in Figure 14a, and the cooling position is shown in Figure 14b. The temperature curve is the average of five points.

#### Table 2. Inhibition Situations

| test | cooling temperature (K) | beginning time (s) |
|------|--------------------------|-------------------|
| 1    | 293.15                   | 100               |
| 2    | 283.15                   | 180               |
| 3    | 273.15                   | 180               |

Figure 13. Schematic of the geometry.

Figure 14. Battery boundary condition: (a) battery heating position and temperature measuring point; (b) battery cooling position (the cooling part is indicated as the blue area).

Equation 2 also applies to the calculation of heat production in this part

$$k_{\text{pe}}(T, c_{\text{pe}}) = A_{\text{pe}} \exp\left(-\frac{E_{\text{a,pe}}}{RT}\right)$$  \hspace{1cm} (4)

where $k_{\text{pe}}$ is the reaction kinetic parameter; $T$ is the temperature; $c_{\text{pe}}$ is a dimensionless number related to the concentration; $\alpha$ is the percentage of the reaction. More detailed parameters can be found in Table 1. In the positive electrolyte runaway reaction, the exothermic process has two maximum values, which can be shown by this side reaction.\textsuperscript{26,27}

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Notes
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