

Article

Investigation of Thermal Runaway Propagation Behavior of 280 Ah LiFePO₄ Battery and Pack Under Overheating Conditions

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Abstract

The extensive utilization of LiFePO₄ (LFP) batteries in energy storage facilities has been impeded by the inherent property of thermal runaway (TR). This study examines the TR propagation characteristics of 280 Ah LFP batteries and their module through the application of dual-side heating to trigger TR. Experimental investigations on single battery TR reveal that the timing and temperature at which the battery safety valve opens exhibit stochastic behavior. Moreover, a correlation is observed between the time required for the safety valve to open and the average surface temperature of the battery, with longer durations corresponding to higher temperatures. Surface temperature variations in batteries manifest in three primary phenomena: temperature decline, abrupt temperature spikes, and peak temperatures. In TR experiments involving packs, it is depicted that temperature signals can detect internal development processes earlier than smoke signals when TR initiates within the module. Heat transfer within batteries of the same sub-module primarily occurs through conduction, exhibiting an average heat transfer fraction of 25.8%. These findings hold significant implications for enhancing early detection systems for TR in both batteries and modules.

Keywords: lithium-ion battery; thermal runaway; battery pack; gas diffusion behavior

1. Introduction

Electrochemical energy storage has been extensively utilized in energy storage systems and power stations due to its advantages, such as high charge or discharge rates, rapid response, cost-effectiveness, and ease of distributed deployment [1]. LFP batteries, in particular, are known for their superior energy density, extended cycle life, and enhanced safety, making them ideal for systems with strict safety requirements [2,3]. However, under non-standard operating conditions like thermal, electrical, or mechanical abuse, LFP batteries are prone to TR, jeopardizing the safety of the entire module [4,5]. TR, as an intrinsic characteristic of LFP batteries [6], poses critical safety concerns, accompanied by the generation of heat and flammable or explosive gases during the process. Hence, further investigation into the evolution of physical fields during TR, such as electrical, gaseous, thermal, mechanical, and smoke-related parameters, is of significant importance for ensuring the safe operation of battery systems.

Several studies have explored the TR characteristics of batteries using different abuse tests, including external heating [7], overcharging [8], over-discharging [9], nail



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penetration [10], internal short circuits and internal external circuits [11]. These investigations revealed that the primary factor leading to TR was internal self-heating resulting from these abusive conditions, where the heat produced was not effectively dissipated [12], culminating in TR initiated by increased temperatures. External heating is a commonly utilized technique in abuse testing due to its convenience and control ability. However, existing research has predominantly focused on one-sided heating as the primary methodology. Batteries do not operate in isolation under real-world conditions. Within a pack, multiple batteries are arranged in proximity, and during operation, each battery generates heat. In typical working environments, individual batteries experience heating from both sides. This bilateral heating results in a relatively rapid increase in temperature. The intensity of the temperature rise during the TR process is notably greater [13]. Therefore, further investigation into the TR propagation characteristics of batteries under conditions of bilateral overheating is warranted.

Due to its complex internal structure, advanced experimental equipment and techniques are required to monitor the internal changes during TR. To better understand the internal temperature during TR, Ouyang's group [14–17] employed thermocouples implanted within the battery between jelly rolls to investigate in-cell rather than the lateral surface, temperature. Fransson et al. [18] used high-speed X-ray imaging to capture, for the first time, an internal sidewall breach process caused by nail penetration and analyze the speed of layer delamination. Meanwhile, from the perspective of TR energy, Zhang et al. [19–21] developed a TR model using 280 Ah LFP batteries to describe internal thermal characteristics, proposing trigger energy as a key feature for evaluating and characterizing TR under different thermal abuse conditions. Song et al. [22,23] extensively investigated the critical conditions for TR in 40 Ah batteries, focusing on temperature and energy aspects through experiments. They elucidated the TR behavior under adiabatic conditions and determined the critical temperature and energy levels for TR at various heating rates. Feng et al. [24–26] studied the TR mechanism of lithium-ion batteries based on thermal analysis databases and found that the total heat generated by internal short circuits during TR accounts for only a small portion. In conclusion, a well-established theory has emerged concerning the variation in physical quantities within the battery during TR. Under actual operating conditions, monitoring external physical parameters (temperature, voltage, and exhaust behavior) during TR and analyzing their temporal changes are crucial for safe operation of energy storage systems. As the functions of energy storage power stations continue to evolve, single-capacity batteries increasingly fail to meet market demands. Several researchers have established experimental platforms to investigate the TR behavior of batteries with varying capacities [27–30]. Their findings indicated that the external physical properties of batteries exhibited distinct variation patterns during TR progression.

In energy storage facilities, LFP batteries are arranged in sub-modules and modules. However, it is crucial to consider the cumulative impacts and potential hazards associated with their deployment [31,32]. Two primary concerns influence the secure functioning of these modules: heat dissipation preceding TR and gas dissemination subsequent to safety valve activation. Song et al. [33–35] further investigated the thermal propagation behavior of 280 Ah LFP battery modules, finding that thermal propagation only occurred at 100% state of charge (SOC), with 75% of the energy used to heat the battery itself. Qin et al. [36–39] probed the gas diffusion inside the battery pack with different venting areas and found that the reason for the battery pack explosion was H_2 and C_2H_4 . Wei et al. [15,40] compared gas emissions from LFP and NCM batteries, finding that LFP batteries primarily emit CO_2 and H_2 , while NCM batteries emit CO_2 and CO. However, gaps remain in the current research. A systematic framework for investigating heat trans-

fer and smoke exhaust behavior within modules has yet to be established in practical application scenarios.

In conclusion, while there is a foundational body of research on the TR mechanism of LFP batteries, there is a scarcity of studies on the propagation process of TR under bilateral heating conditions. Additionally, limited analyses exist on the evolution of external physical parameters of batteries over time during TR, particularly within actual modules. Therefore, this paper focuses on discussing the TR propagation characteristics among individual batteries and packs, addressing the issues identified earlier. To ensure experimental accuracy and mitigate the impact of battery variations on results, five single-battery TR experiments were conducted for both qualitative and quantitative analysis of TR characteristics. The primary tasks included establishing a TR experimental platform, describing the experimental phenomena of TR, analyzing changes in external physical parameters during the TR process, and summarizing the failure mechanisms of cells and modules. Analyzing temperature, voltage, and exhaust behavior over time is vital for improving early safety warnings for energy storage systems.

2. Experiments

Energy storage power stations are essential elements of contemporary energy systems, playing a crucial role in facilitating the transition to sustainable development. An integrated energy storage system combines essential components, including individual cells, modules, thermal management systems, safety protection systems, and other supporting structures, into standardized cabinets. Its structural design aims for high energy density, enhanced safety, exceptional reliability, and ease of maintenance. The primary objective is to ensure the safe operation of modules, as illustrated in Figure 1. The experimental research focuses on a single cell and pack. Figure 1a demonstrates the arrangement of batteries interconnected in series and parallel to create modules. These modules are further organized within cabinets using specific connection techniques to establish an energy storage power station. Therefore, the main object of this experiment is the single cell and its module.



Figure 1. Battery, pack, and energy storage power station. (a) Battery, pack, and energy storage power station inter-structure; (b) Battery structure; (c) Pack structure.

2.1. Experimental Subject

The experimental setup utilized LF280K-type single batteries, which are employed in energy storage systems. The basic physical parameters are detailed in Table 1. Where S_1 represents the battery volume, m_1 denotes the net weight, M_c stands for the cathode material, M_a represents the anode material, C_1 indicates the nominal capacity, and V_1 signifies the voltage.

Table 1. Basic parameters of the battery.

Item	Value
S_1 (mm ³)	207.5 mm × 173.7 mm × 72.0 mm
m_1 (kg)	5.42
M_c	LiFePO ₄
M_a	graphite
C_1 (Ah)	280
V_1 (V)	3.2

As shown in Figure 1b, the battery primarily consists of an aluminum shell, cell core, plastic thermal insulation shell, electrode materials, and electrolyte. The battery module used is 1P52S, composed of four sub-modules connected in series, with each sub-module containing 13 single cells in series. The basic parameters are listed in Table 2, where S_2 denotes the module's volume, m_2 indicates the total mass, L represents the minimum spacing between sub-modules, C_2 refers to the nominal capacity, V_2 signifies the total voltage of all batteries, P_{cd} is the maximum charging and discharging power, and M_u specifies the material of the casing. The primary components of the pack, as depicted in Figure 1c, comprise the upper cover, pressure balance valve, cooling plate, sub-modules, aerogels, sealing strip, and support structure. The upper cover is securely fastened to the sealing strip using bolts. A specific gap exists between the top of the sub-module and its four sides.

Table 2. Basic parameters of the module.

Item	Value
S_2 (mm ³)	1160 mm × 810 mm × 245 mm
m_2 (kg)	330
L (mm)	3.5
C_2 (Ah)	280
V_2 (V)	166.4
P_{cd} (KW)	23.3
M_u	thermoplastic material

2.2. Experimental Setup and Procedures

Before commencing the experiment, it is essential to ascertain the state of the battery, the surrounding environment, and the method for triggering TR. Single cell and module experiments are conducted independently, employing distinct experimental techniques. Single cell tests are performed in closed containers, whereas module TR experiments are executed in a relatively open environment, utilizing a suitably sized fume hood to capture the fumes and gas mixtures produced by the module. The specific operational methods are outlined as follows.

2.2.1. Setup

The laboratory environment was maintained at a temperature of 25 ± 5 °C and a relative humidity of $50 \pm 5\%$. The characteristics of thermal runaway (TR) propagation are

exacerbated by an increase in state of charge (SOC). The duration of TR propagation in a pack at 100% SOC is 70 s shorter than that observed in a pack at 70% SOC. Notably, a cell at 50% SOC does not undergo TR [41], whereas the repercussions of TR in a battery pack at 100% SOC are considerably more severe [42,43]. Consequently, all experimental subjects must undergo a minimum of two charge–discharge cycle adjustments, maintaining 100% SOC and stability for a period ranging from 1 to 8 h prior to testing.

2.2.2. Single Cell Experiments

In practical engineering applications, the performance of an individual cell is influenced by manufacturing variations and environmental factors. Even batteries from the same production batch may exhibit slight differences in internal parameters. These variances tend to magnify over time, impacting the collective performance of the pack. To ensure the accuracy of TR experiments, experiments were conducted on five identical batteries of the same type and manufacturer by employing bilateral heating to induce TR. The schematic diagram of the battery TR experimental platform is depicted in Figure 2. A combustion chamber comprising a sealed pressure vessel was used for these experiments, capturing the smoke and mixed gases produced post-TR. Gas chromatography and explosion limit analyzers (Shanghai Nuoxi Instrument Co., LTD., Shanghai, China) were subsequently utilized to analyze the composition of the mixed gases. The experimental platform comprises four main components: the combustion chamber, exhaust system, sampling and analysis system, and data analysis and recording system. This configuration allows for the real-time monitoring and recording of key parameters during TR, including temperature, voltage, heat release rate, and mass variations. The chamber is equipped with explosion-proof observation windows, and a digital camera records real-time image data throughout the TR process.

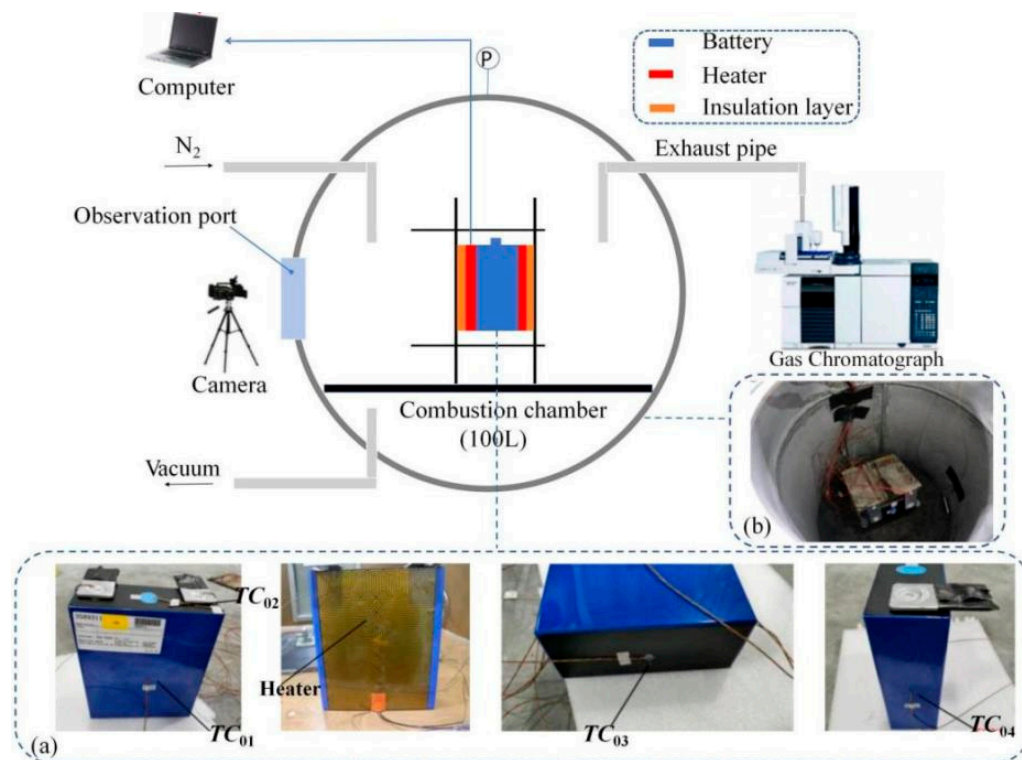


Figure 2. Experimental setup of the battery cell. (a) Thermocouple arrangement diagram; (b) Internal view of the setup.

Heating plates with dimensions of 203.0×152.4 mm were installed on both sides of the cell. A surface heating rate of 4 to 7 °C per-minute was applied to the battery. Additionally, thermal insulation plates were placed on both sides to minimize heat dissipation. Four thermocouples were arranged on the battery surface to monitor temperature changes (Figure 2a): beneath the heater (TC_{01}), near the battery positive terminal (TC_{02}), at the battery bottom (TC_{03}), and on the battery side (TC_{04}). TC_{05} represents the ambient temperature, while V1 denotes the terminal voltage of a single cell. During voltage measurement, the voltage lines are connected to the cathode and anode terminals of the battery, and the circuit remains open throughout the heating process.

2.2.3. The Experiments on the Battery Pack

The pack TR propagation test was conducted in a spacious area. A suitably sized smoke hood was used to collect the smoke and mixed gases produced by the module. Measurement of the chemical heat release rate requires a system comprising a paramagnetic oxygen analyzer (Shanghai Nuoxi Instrument Co., LTD., Shanghai, China), a non-dispersive infrared carbon dioxide and carbon monoxide analyzer (Shanghai Nuoxi Instrument Co., LTD., Shanghai, China), a velocity probe (Chongqing Ode Instrument Co., LTD., Chongqing, China), and a K-type thermocouple (Chongqing Ode Instrument Co., LTD., Chongqing, China). These instruments should be positioned in the exhaust duct of the heat release rate calorimeter to minimize the impact of bends or exhaust devices. Analysis of the vent gas composition should be performed using a Fourier-Transform Infrared Spectrometer with a resolution of at least 1 cm^{-1} and a path length of no less than 2 m (6.6 ft), or an equivalent gas analyzer. Velocity and temperature measurements in the exhaust duct of the heat release rate calorimeter require suitable equipment. The hydrocarbon content in the vent gas should be analyzed using flame ionization detection, and H_2 levels must be evaluated with a palladium–nickel thin–film solid-state sensor. Light transmission in the exhaust duct of the calorimeter should be assessed by utilizing a white light source and a photo detector during the test to calculate the smoke release rate.

The 34# cell is induced into TR through a dual-sided heating method. By regulating the input power of the thin-film heater, a surface heating rate of 4 to 7 °C per minute is attained, with a maximum heating power of 430 W. Within the pack, aerogel functions primarily as a high-efficiency thermal insulator and flame-retardant material. Its essential role is to delay or obstruct the propagation of TR between cells, thereby improving the overall safety of the pack. The module experiment focuses on investigating the TR behavior of the pack under actual usage conditions. Aerogel is positioned between all batteries, and the key parameters are presented in Table 3, where T_a is the thickness, ρ_a is the density, c_{pa} is the specific heat capacity, and k is the heat transfer coefficient.

Table 3. Basic parameters of the aerogel.

Item	Value
T_a (mm)	1.3 ± 0.1
ρ_a ($\text{kg} \cdot \text{m}^{-3}$)	195
c_{pa} ($\text{J} \cdot \text{kg}^{-1} \cdot ^\circ\text{C}^{-1}$)	700
k ($\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$)	$25^\circ\text{C}, \leq 0.030; 100^\circ\text{C}, \leq 0.035$

A total of 52 batteries were ranged within the pack. The aerogel's heat insulation effect allows for the collection of temperature data from three adjacent batteries within the sub-module. Given the relatively large lateral distance between the sub-modules, TR is unlikely to propagate between the battery packs; thus, only the temperature information from adjacent batteries is gathered. K-type thermocouples were employed to monitor

temperature variations on each battery surface. The placement of the thermocouples is illustrated in Figure 3. V_2 denotes the total voltage of the battery module. When subjected to heat, the circuit opens, and the voltage measurement lines are connected to the positive and negative terminals of the pack.

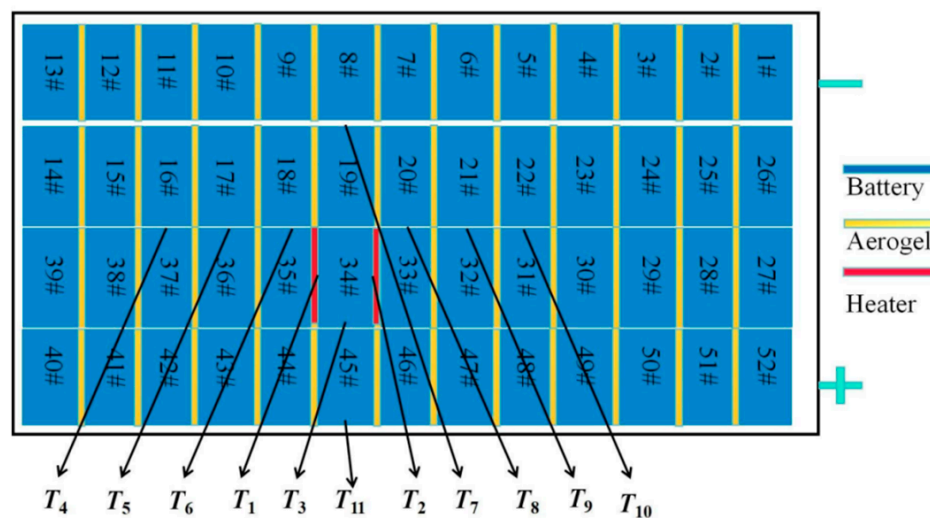


Figure 3. Thermocouple arrangement diagram.

3. Results and Discussion

The experimental phenomena resulting from conducting cell and pack experiments separately exhibit differences. Single battery experiments investigate exhaust and open valve behavior, gas detection, and rules governing temperature and voltage changes. Module experiments concentrate on studying the propagation behavior of thermal runaway (TR), analyzing exhaust behavior, temperature and voltage change patterns, heat transfer ratios, and smoke release. Ultimately, a failure mechanism for the operation of single batteries and modules is summarized based on the occurrence and transmission characteristics of TR.

3.1. Battery Experimental Results

3.1.1. Exhaust and Open Valve Behavior

Figure 4 illustrates the phenomena occurring during the TR. Under overheating conditions, the battery did not combust. From the onset to the conclusion of TR, the process can be divided into four stages:

(I) Initiation of TR: Under the influence of the heating plate, the battery temperature continuously rises. The electrolyte evaporates, accompanied by other chemical reactions, leading to the continuous generation of mixed gases. These gases accumulate at the safety valve outlet, resulting in an increase in internal pressure.

(II) Safety valve activation: Due to the internal gas pressure, the safety valve opens instantaneously, releasing a large volume of mixed gases in a brief jet stream. Upon contact with the external environment, the electrolyte in the jet stream cools and forms a white mist.

(III) Continuous gas emission: As the temperature rises and internal reactions persist, TR occurs throughout the battery. Mixed gases continue to be expelled, accompanied by smoke and other solid particles.

(IV) Termination of gas emission: Internal reactions within the cell cease, with only a small amount of mixed gas escaping until the TR reaction concludes. Following the TR reaction, the cell sustained severe damage.

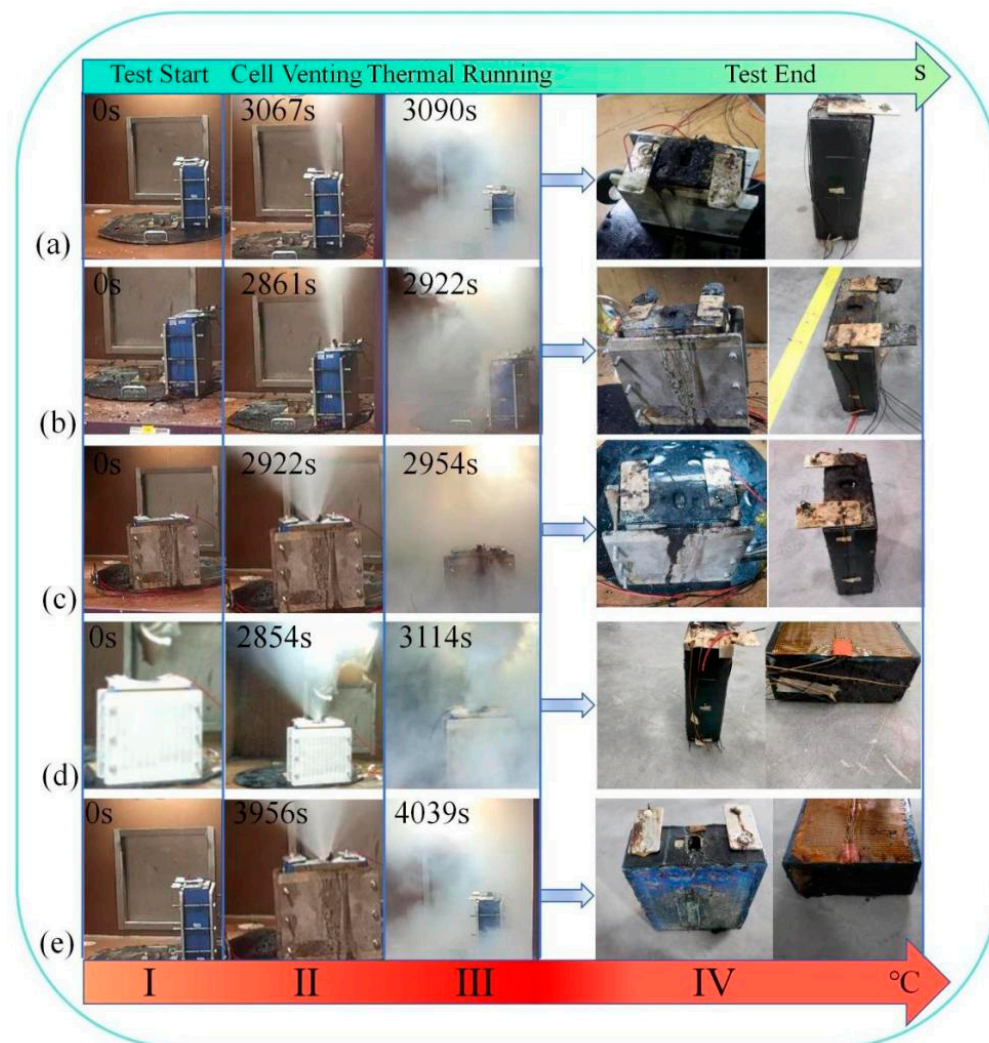


Figure 4. TR experimental phenomena of single batteries. (a–e): Battery-1~Battery-5.

A lot of black particles remained at the safety valve, and varying degrees of burn and corrosion marks were observed at the positive and negative electrode interfaces. The surface was coated with electrolyte, attributed to the lithium salt LiPF_6 used in the experimental battery. The generated PF_6 exhibits strong corrosiveness, causing varying degrees of corrosive damage to both the internal and external components of the cell during TR.

The opening times of the safety valves, based on the audio and video signals, and temperature data for each cell are shown in Figure 5. Variability in the opening times of the safety valves arises from manufacturing discrepancies and the superimposition of internal side reactions, resulting in a degree of randomness. For Batteries 1–4, the opening times predominantly cluster between 2800 and 3000 s (s), with the average surface temperature at the moment of valve opening ranging from 140 to 160 °C. In contrast, the safety valve of battery-5 opens significantly later, exhibiting a delay of approximately 900 s. The data indicate that longer safety valve opening times correspond to higher average surface temperatures. This reflects an extended duration of thin-film heater operation. When the temperature attains a specific critical threshold, a series of chain reactions occur internally, generating heat and releasing gas. This process may cause the safety valve to open randomly, potentially triggering TR. At this juncture, if the battery continues to draw energy from external sources, its safety equilibrium will ultimately be compromised.

Consequently, the surface temperature of the cell prior to the activation of the safety valve is crucial for the early warning of TR.

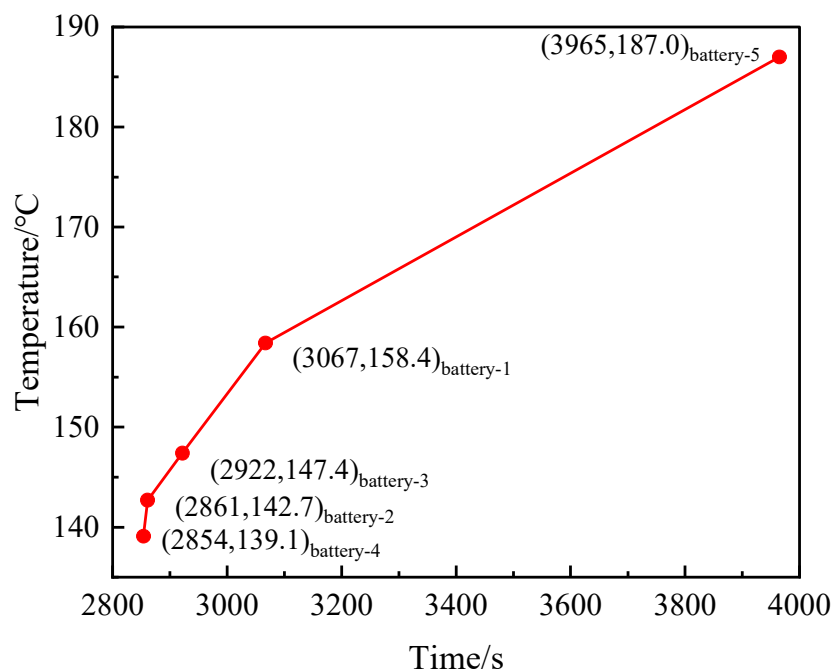


Figure 5. Safety valve opening time and average temperature.

3.1.2. Gas Detection

Following the TR experiments, the gases from the sealed containers underwent extraction and analysis via gas chromatography. The results of the five experiments consistently revealed similar gas components with minimal volume fraction errors. Therefore, the average value of experiment is selected as the result in Table 4. Notably, the predominant gases released upon opening the safety valve included H₂, CO₂, CO, CH₄, and C₂H₂. H₂ emerged as the most abundant gas, constituting 48.013% of the total gas volume, followed by CO₂ at 27.325%. Hydrogen has a small molecular weight and is absent in atmospheric composition. Therefore, its significant presence in the vent gas makes it a valuable indicator for safety alerts.

Table 4. Volume fraction of each gas component.

Item	Measure	Conc. (%)
1	CO	11.191
2	CO ₂	27.325
3	H ₂	48.013
4	CH ₄	6.404
5	C ₂ H ₂	0.107
6	C ₂ H ₄	3.296
7	C ₂ H ₆	1.326
8	C ₃ H ₆	0.948
9	C ₂ H ₈	0.321
10	C4(Total)	0.704
11	C5 (Total)	0.142
12	C6 (Total)	0.005
13	C7 (Total)	0.003
14	C ₆ H ₆	0.014
15	C ₄ H ₈ O ₃	0.201
Total	/	100.000

3.1.3. Temperature and Voltage

The surface temperature and voltage of cells are critical parameters for evaluating their internal reaction processes and heat generation characteristics. As shown in Figure 6, the temperature measurement points and voltage variation curves of the five batteries are presented. where V1 is the cell voltage, t is the time, and A is the coordinate of the peak of the curve.

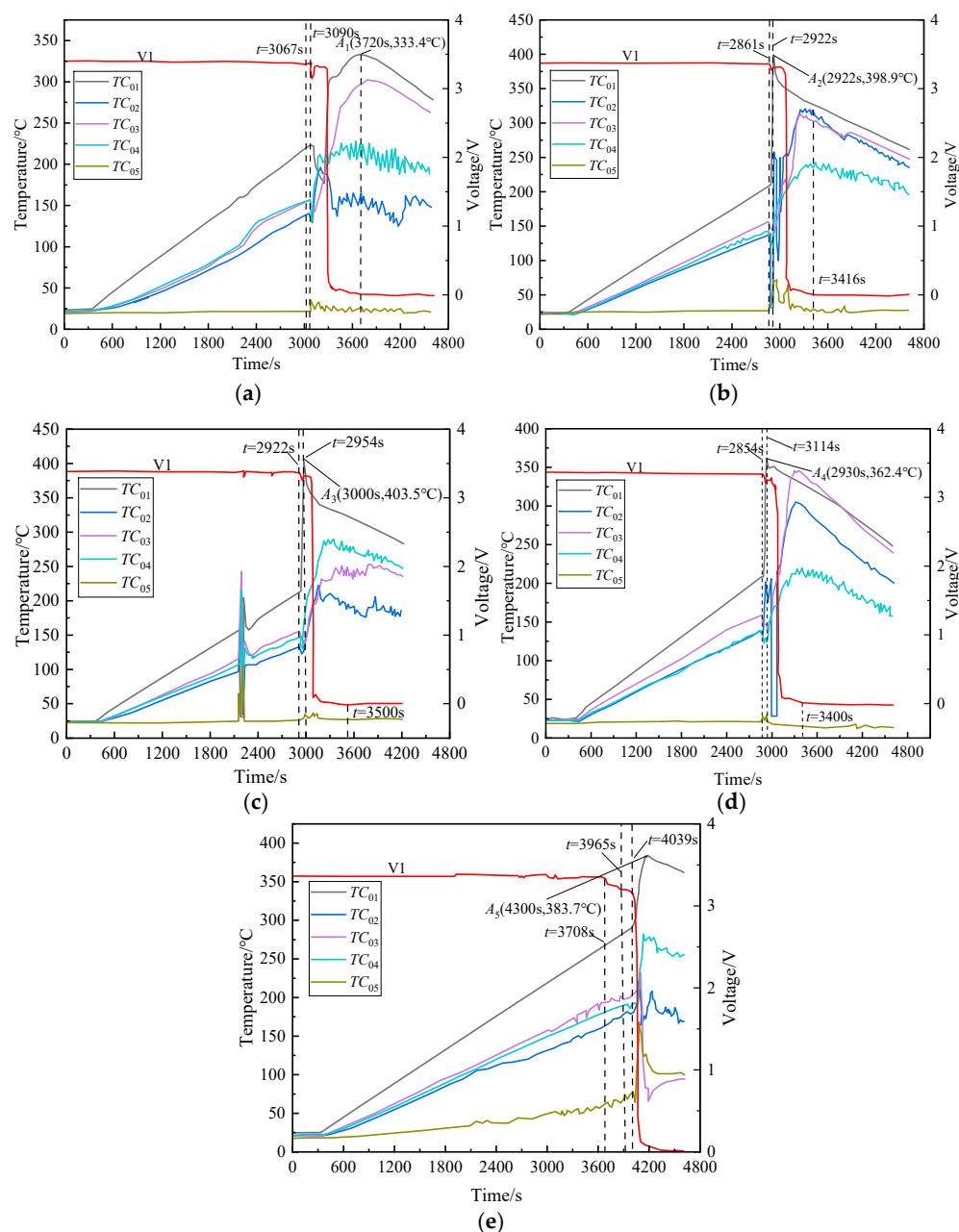


Figure 6. Variation curves of battery temperature and voltage measurement points. (a–e): Battery-1 to Battery-5.

As depicted in Figure 6, the general pattern of temperature and voltage remains consistent. As illustrated in Figure 6a,c, the surface temperatures of battery-1 and battery-3 exhibited significant fluctuations at 2400 (s). However, this variability did not alter the overall trend. After a period, the temperatures returned to normal levels, a phenomenon attributed to the manufacturing differences among the cells. Upon reaching approximately 3000 to 4000 s, the safety valve discharged smoke, coinciding with an abrupt surge in ambient temperature. Upon opening the safety valve, three sequential phenomena were

observed in the surface temperature: (I) A transient temperature drop occurs, followed by a brief temperature increase, and ultimately a decrease in surface temperature. These observations can be attributed to the combined effects of the Joule–Thomson effect [44,45], high-temperature eject, and the heat dissipation by the electrolyte, leading to the apparent surface temperature drop. The temperature drops on the heating surface and side is more pronounced. This is attributed to enhanced electrolyte evaporation and heat absorption near the heat source, as well as a higher boiling convection heat transfer coefficient. Additionally, the temperature at TC_{02} fluctuates significantly due to its proximity to the safety valve. This fluctuation is influenced by the jet state when the safety valve opens, leading to a sudden change in ambient temperature, with a maximum fluctuation of 227.0 °C. (II) The rapid increase in temperature, known as the critical point, marks the onset of a sharp rise in temperature as the initial temperature for TR. The timing of this occurrence varies across different surfaces. The heating surface exhibits the highest temperature increase and rate due to the larger surface area and the presence of more electrolytes compared to other surfaces. (III) As the temperature reaches its peak, internal TR within the battery occurs. The surface temperature progressively increases to its maximum value before gradually declining as the internal reaction concludes. The voltage began to decrease upon the opening of the safety valve, exhibiting a temporary recovery before sharply declining thereafter. This decline in voltage signified the malfunction of both cells attributed to a transient rebound in the parallel voltage of the two cells.

Table 5 defines the parameters used to quantify TR characteristics across three phases: t_{sv} is the safety valve opening time, ΔTC_{05} is the ambient temperature change value, ΔTC_{01} to ΔTC_{04} is the temperature variation values at each stage, R_{01} to R_{04} is the rate of temperature change, t_{dv} is the time when the voltage starts to drop, and t_{d0} is the time it takes for the voltage to start dropping until it falls to zero.

Table 5. Temperature values and rate of change for each phenomenon.

Battery		Battery ₁	Battery ₂	Battery ₃	Battery ₄	Battery ₅
t_{sv} (s)		3067	2861	2922	2854	3965
ΔTC_{05} (°C)		13.9	45.1	10.2	7.8	10.1
I	$\Delta TC_{01}/R_{01}$ (°C/°C·s ^{−1})	38.6/0.22	−/−	−/−	−/−	−/−
	$\Delta TC_{02}/R_{02}$ (°C/°C·s ^{−1})	11.2/0.28	−/−	10.1/0.31	−/−	−/−
	$\Delta TC_{03}/R_{03}$ (°C/°C·s ^{−1})	25.1/1.04	16.1/0.21	17.9/0.25	23.8/0.29	−/−
	$\Delta TC_{04}/R_{04}$ (°C/°C·s ^{−1})	14.0/0.24	19.8/0.38	18.0/0.64	16.8/0.28	−/−
II	$\Delta TC_{01}/R_{01}$ (°C/°C·s ^{−1})	118.6/2.5	187.8/3.07	189.4/3.0	153.6/3.49	93.0/0.59
	$\Delta TC_{02}/R_{02}$ (°C/°C·s ^{−1})	66.9/0.65	179.1/5.2	99.6/0.47	68.8/1.96	43.8/0.63
	$\Delta TC_{03}/R_{03}$ (°C/°C·s ^{−1})	126.5/0.39	77.8/0.57	60.1/0.56	76.2/0.63	26.2/0.41
	$\Delta TC_{04}/R_{04}$ (°C/°C·s ^{−1})	76.8/1.05	45.1/1.87	73.3/1.52	45.8/1.01	92.9/1.34
III	$\Delta TC_{01}/R_{01}$ (°C/°C·s ^{−1})	333.4	398.9	403.5	362.4	383.7
	$\Delta TC_{02}/R_{02}$ (°C/°C·s ^{−1})	196.0	320.7	225.7	305.3	224.4
	$\Delta TC_{03}/R_{03}$ (°C/°C·s ^{−1})	302.2	312.3	252.4	345.5	232.2
	$\Delta TC_{04}/R_{04}$ (°C/°C·s ^{−1})	211.9	242.2	289.8	220.7	281.9
t_{vd} (s)		3090	2861	2922	2854	3708
t_{d0} (s)		630	549	578	546	900

Upon activation of the safety valve, the ambient temperature TC_{05} increased by a maximum of 45.1 °C. Variability in temperature reduction resulting from internal battery reactions was observed, with surface manifestations being non-uniform and displaying a degree of randomness. The maximum temperature decrease and rate of temperature decline due to cell reactions exhibited variability without a discernible pattern. Fluctuations in TC_{02}

levels following safety valve release were noted, contributing to inconsistent temperature reduction patterns. The maximum observed surface temperature decrease was $38\text{ }^{\circ}\text{C}$, with a maximum temperature decline rate of $1.04\text{ }^{\circ}\text{C/s}$. The onset of TR is characterized by a sudden increase in temperature, with the highest temperature rise value and rate observed at TC_{01} due to the heat source, reaching a maximum of $189.4\text{ }^{\circ}\text{C}$ and $3.49\text{ }^{\circ}\text{C/s}$, respectively. The area where electrolyte accumulates during TR, namely the bottom surface, exhibits more consistent temperature rise values and rates, peaking at $92.9\text{ }^{\circ}\text{C}$ and ranging from 1.01 to $1.87\text{ }^{\circ}\text{C/s}$. Throughout the entire TR heat transfer process, the cell's heated surface reaches a maximum temperature of $403.5\text{ }^{\circ}\text{C}$, while the remaining surface peaks at $345.5\text{ }^{\circ}\text{C}$. Voltage fluctuations are observed concomitantly with the TR progression: the voltage starts declining prior to and following the opening of the safety valve. Notably, the delay in opening the safety valve results in increased heat absorption, prolonging the required time, with a maximum duration of 900 s .

3.2. Pack Experimental Results

3.2.1. Exhaust Behavior

As illustrated in Figure 7, the TR experiment of the pack did not result in flame combustion, explosion, or debris scattering during the propagation process.

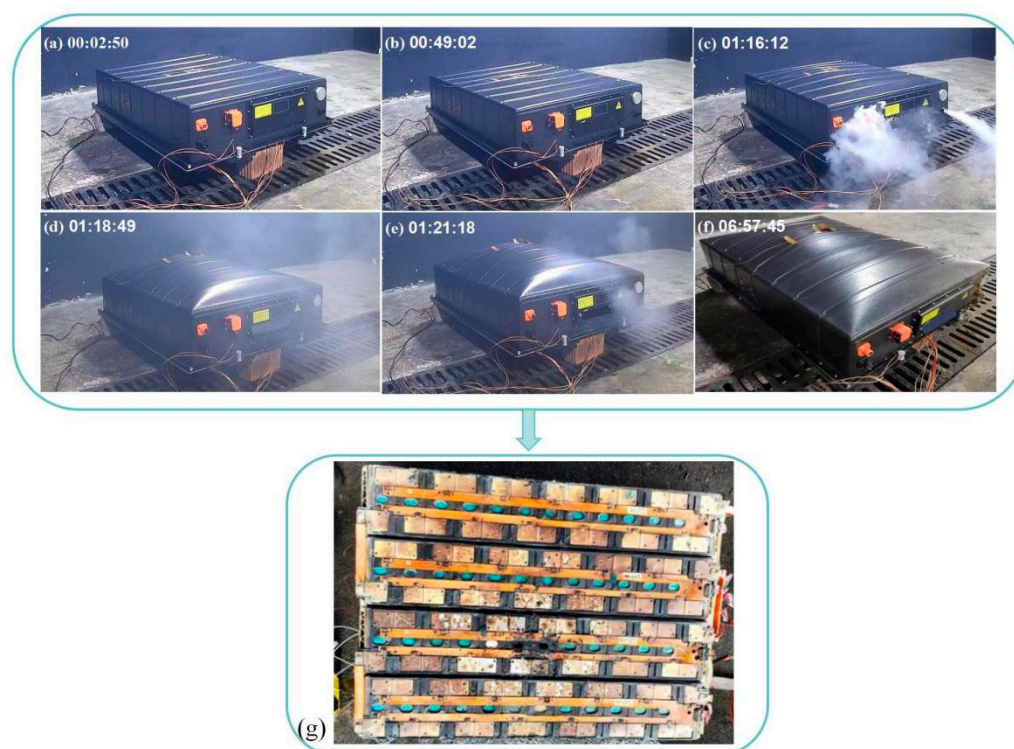


Figure 7. Experimental phenomena of the module. (a): heater startup time; (b): the slight expansion time of the upper cover; (c): flue gas release time; (d): the upper cover continues to expand for a while; (e): the continuous release time of flue gas; (f): experiment end time; (g): the internal situation after the experiment.

In Figure 7a, at $2\text{ min } 5\text{ s}$, the heaters on both sides were activated simultaneously to heat the two primary surfaces of the battery 34#. Figure 7b shows that after $46\text{ min } 12\text{ s}$, the upper cover begins to exhibit slight expansion, indicating that the safety valve has opened. As the exhaust volume continues to increase, the internal pressure of the pack rises correspondingly. The upper cover deforms and expands in response to this pressure. When the internal pressure surpasses a specific threshold, the pressure relief valve activates, releasing smoke and mixed gases, as depicted in Figure 7c. Subsequently,

the heater is turned off. At 68 min 49 s into the experiment, the upper cover exhibited significant swelling, accompanied by the release of a small amount of gas. The safety valve of battery 33# opened, and 2 min 37 s later, smoke and mixed gases were expelled, resulting in an increased concentration of mixed gases in the environment and widespread diffusion of smoke, as illustrated in Figure 7d. By 81 min 18 s, smoke was observed to be expelled again, with the upper cover continuing to swell, as shown in Figure 7e. The experiment concluded without any other cells experiencing TR; the deformation of the upper cover gradually decreased, and no further smoke was released, as depicted in Figure 7f. In total, two batteries within the pack underwent TR, while the remaining batteries remained unaffected due to the protective properties of the thermal insulation aerogel. Within the pack, the safety valves of batteries 33# and 34# activated, leading to electrolyte overflow and subsequent corrosion and burning of adjacent batteries and components. The TR was confined to the batteries within the sub-modules and did not propagate to other sub-modules, as illustrated in Figure 7g, which presents the internal diagram of the module following the experiment.

3.2.2. Temperature and Voltage

Figure 8 illustrates the trend graphs of temperature measurement points alongside voltage changes, where T is the battery surface temperature, t is the time, and $dT \cdot dt^{-1}$ is the rate of temperature change.

As depicted in Figure 8a, during the initial phase of the experiment, the surface temperatures across various sections exhibited a linear increase due to the influence of the heating plate. At 47 min 54 s, the battery safety valve activated, coinciding closely with the observed experimental time. Following this event, the temperature experienced a notable decrease. This reduction can be attributed to the Joule–Thomson effect and the combined influence of high-temperature eject and electrolyte, which collectively dissipated some heat, leading to a decline in the surface temperature of. At this juncture, the average surface temperature of the battery was recorded at 271.5 °C. After heating for 28 min 48 s, the temperature rise rates for each surface reached their maximum values, recorded at 1.45 °C/s, 2.54 °C/s, and 0.59 °C/s, respectively. This data indicates that the cell underwent TR, leading to a sharp increase in surface temperatures until the maximum temperature was attained at 78 min 54 s. The recorded temperatures were 458.5 °C, 581.5 °C, and 309.1 °C, yielding an average of 449.7 °C. This average represents a 39.6% increase compared to the average surface temperature at the time the safety valve opened. Notably, the temperature rise rate and maximum temperature on the surface of T_2 exceeded those on the surface of T_1 , attributed to the battery's asymmetrical internal structure and irregular electrolyte distribution. The entire TR process, from the initiation of heating to the attainment of the maximum temperature, lasted 78 min 54 s. Notably, the interval from the opening of the safety valve to the maximum temperature observed on each surface spanned 31 min, representing 39.2% of the total duration. In contrast, the time from the onset to the conclusion of the TR was merely 3 min 12 s, accounting for 0.41% of the overall process. This observation underscores the rapid progression of TR. Consequently, the limited response time hampers effective intervention. During the heat transfer and accumulation, a secondary peak temperature of 565.2 °C was recorded on the surface of T_1 , attributed to the self-generated heat of battery 33# reheating the T_1 surface for a second time. Subsequently, as heat continued to transfer and diffuse over time, the temperature gradually decreased.

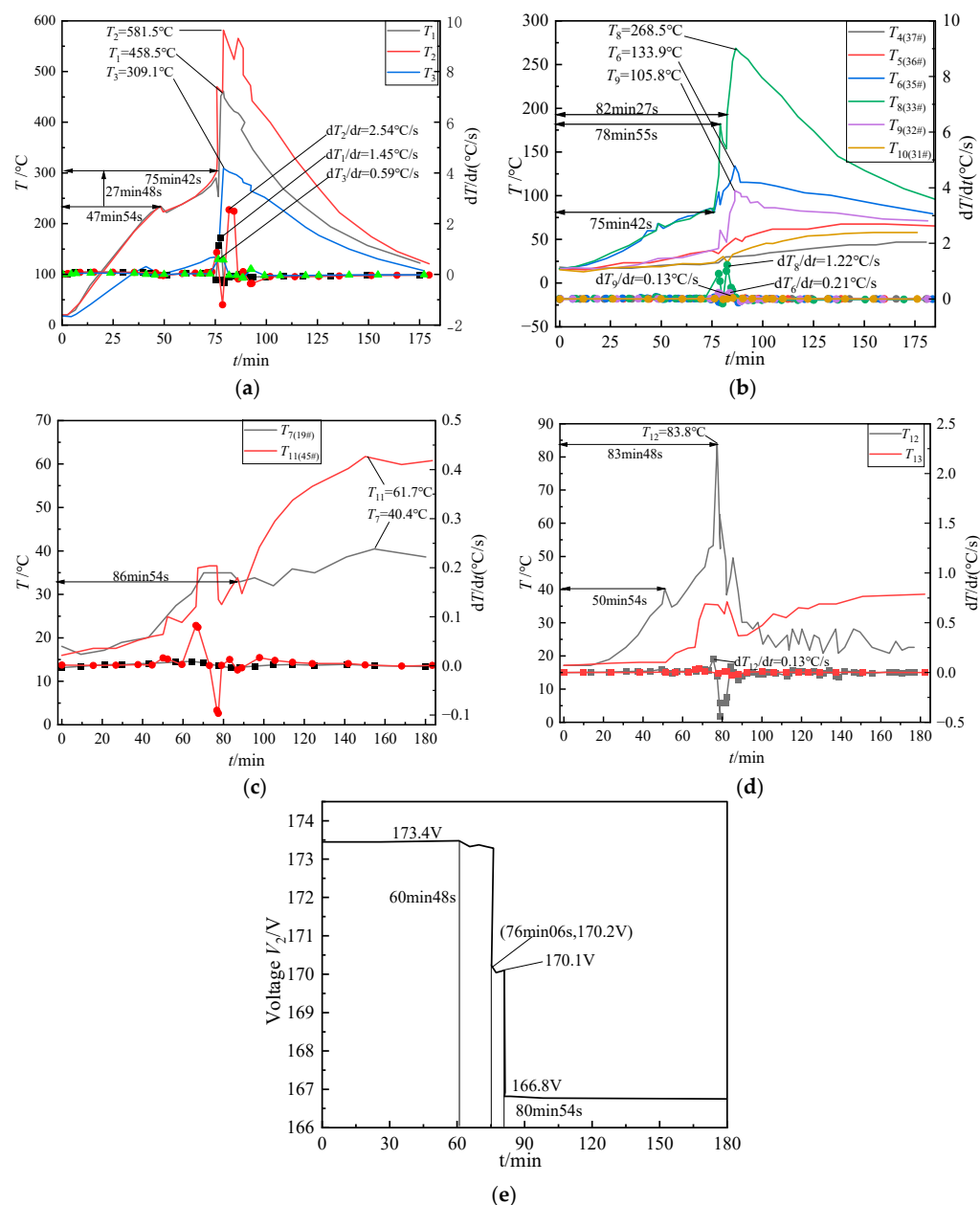


Figure 8. Temperature, temperature rise rate and voltage variation in each thermocouple measurement point. (a): 34#; (b): 1#–37#; (c): 19# and 45#; (d): upper cover; (e): voltage.

To investigate the heat transfer between batteries following the occurrence of TR within the module, the temperature variations on the adjacent sides of the batteries are illustrated in Figure 8b. Battery 34# acts as a heat source for the neighboring batteries. Prior to the onset of TR, the temperatures of the surrounding batteries gradually increase, representing the normal heating phase. At 75 min 42 s, battery 34# undergoes TR, resulting in a significant rise in the temperatures of the adjacent batteries, accompanied by an accelerated temperature increase rate. Heat is transferred through both the left and right pathways; due to the temperature differential between T_1 and T_2 , the temperature changes along each side of the pathways differ as well. The temperature change intensity along the right path from T_8 to T_{10} exceeds that of the left path from T_6 to T_4 . Notably, the temperature variation at T_8 of battery 33# is the most pronounced. The safety valve activates at 78 min 55 s, allowing exhaust to dissipate some heat. Following a brief temperature decline, the rate of temperature increase surpasses 1°C/s at 82 min 27 s, peaking at 1.22°C/s . This indicates that battery 33# has undergone TR, with its temperature reaching a maximum of

268.5 °C, which represents a 13.1% difference compared to the side temperature of battery 34#. This discrepancy arises from varying heating rates, aligning with the findings presented in reference [20]. A higher heating rate correlates with a more vigorous TR reaction and a greater maximum temperature. Along the left path (T_6 to T_4), battery 35# exhibited the most significant temperature change at T_6 . Its maximum temperature rise rate was 0.13 °C/s, and its peak temperature was 133.9 °C. No TR was observed.

Heat transfer between sub-modules primarily occurs through convection and radiation. Figure 8c illustrates the variation in characteristic temperatures T_7 and T_{11} for batteries 19# and 45#, respectively. The heat transfer efficiency between the sub-modules is limited. The temperature begins to rise after 86 min 54 s, at which point the thermal runaway process within the module has largely concluded, with maximum temperatures recorded at 61.7 °C for the 19# battery and 40.4 °C for battery 45#. Additionally, T_{11} exceeds T_7 , a phenomenon attributed to battery 45#'s confinement within a smaller space, which facilitates greater heat accumulation.

The upper cover of the pack is the first component to interact with the mixed gas. Temperature changes within this cover can provide direct insights into the internal development processes, as illustrated in Figure 8d. The temperature T_{12} at the top of the upper cover and the temperature T_{13} at its side are of particular interest. The temperature increase at T_{13} is relatively minor, with the rate of increase effectively remaining at 0. The maximum recorded temperature is 35.6 °C, which shows no significant deviation from the ambient temperature. In contrast, the temperature variation at the top of the upper cover is more pronounced. The first temperature peak occurs at 50 min 54 s, which lags behind the safety valve opening time of battery 34# by 6.2%. The second temperature peak is observed at 83 min 48 s, trailing the safety valve opening time of battery 33# by 6.4%, with a maximum temperature of 83.8 °C. This represents an increase of approximately 3.3 times compared to the initial temperature. Due to the specific distance between the upper cover of the module and the safety valve, the hysteresis error remains within an acceptable range. Consequently, the temperature measured at the top of the upper cover can effectively function as a critical signal for the early warning of thermal runaway events occurring within the module.

The overall voltage variation in the module is illustrated in Figure 8e. The total voltage of the module measures 173.4 V, comprising 52 individual batteries connected in series, each with a voltage of 3.3 V. During the initial heating phase, the total voltage remained relatively stable. However, at 60 min 48 s, a decline in voltage was observed. This coincided with the opening of the safety valve of battery 34#, which resulted in internal damage. At 76 min 6 s, the voltage plummeted to 170.1 V, reflecting a decrease of 3.3 V. The temporal discrepancy compared to the maximum surface temperature was 3.5%, indicating that battery 34# had experienced complete failure. At this juncture, the safety valve of battery 33# was activated, resulting in a continued decline in total voltage. At 80 min 54 s, the voltage experienced a rapid decrease to 166.8 V, reflecting a reduction of 3.3 V. The temporal discrepancy relative to the maximum surface temperature was measured at 3.4%, marking the failure of the 33# battery. Throughout the voltage variation process, the voltage began to decline prior to the activation of the safety valve. It subsequently fell to a level where the deviation between the voltage of a single cell and the termination point of thermal runaway was also 3.4%. This observation indicates that the surface voltage can effectively signal the progression of changes associated with TR.

3.2.3. Heat Transfer Fraction

TR occurs within the internal cells of the pack. The primary mechanisms of heat transfer among the batteries include heat conduction, convection, and radiation, as illustrated in Figure 9. Lateral heat transfer between the batteries occurs primarily through

heat conduction, while longitudinal heat transfer is facilitated by both heat convection and radiation. These findings align with the conclusions presented in references [46,47]. The theoretical heat transfer value can be derived from the fundamental heat transfer formula, as expressed in Equation (1). However, during the actual process, the battery safety valve opens, releasing high-temperature mixed gas that removes a portion of the heat while allowing it to accumulate within the module. In this study, Formula (2) is employed to calculate the heat value.

$$\begin{aligned} q_{\text{conduction}} &= -\lambda \frac{(T_1 - T_2)}{\delta} \\ q_{\text{convection}} &= h(T_1 - T_2) \\ q_{\text{radiation}} &= \frac{\sigma(T_1^4 - T_2^4)}{\frac{1-\varepsilon_1}{\varepsilon_1 A_1} + \frac{1}{A_1 F_{12}} + \frac{1-\varepsilon_2}{\varepsilon_2 A_2}} \end{aligned} \quad (1)$$

where q represents the heat flow density, λ is the thermal conductivity, δ is the thickness, h is the surface heat transfer coefficient, σ is the Stefan–Boltzmann constant, ε is the surface emissivity, A is the surface area, and F is the viewing angle coefficient.

$$Q = cM\Delta T \quad (2)$$

where Q denotes the heat energy in joules (J); c is the specific heat capacity of the battery, taken as $2090 \text{ J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$; M is the battery mass, taken as 5.42 kg ; and ΔT is the temperature rise in kelvins (K).

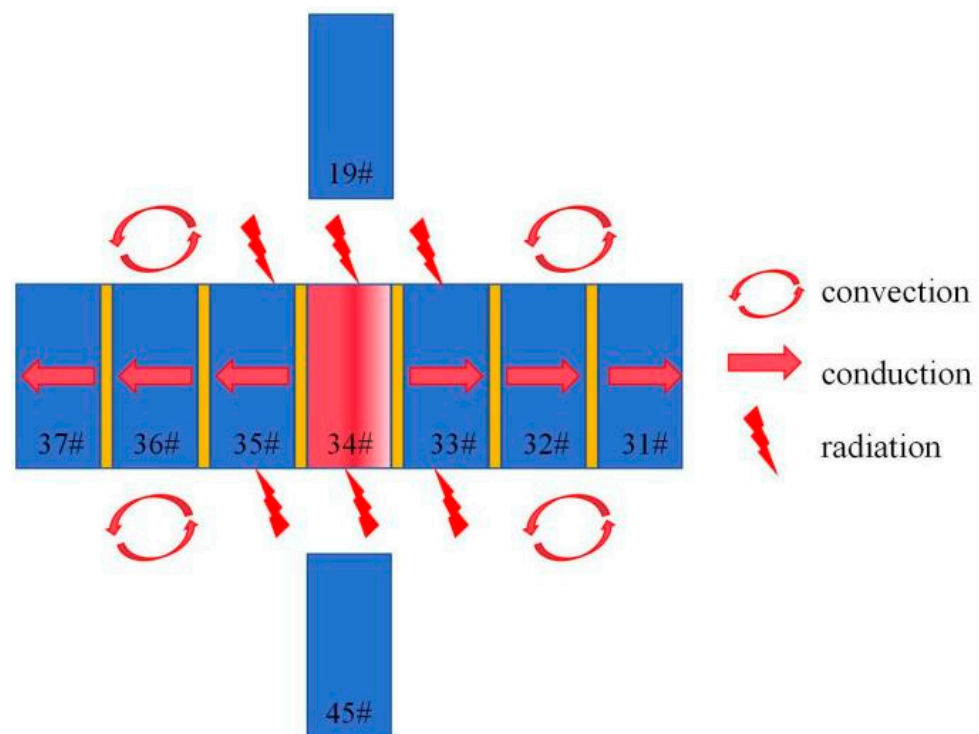


Figure 9. Heat transfer mode inside module.

To accurately quantify heat transfer and consider heat generated by TR, the process is segmented into two stages using the safety valve opening as a reference point. The initial stage, labeled as t_{s1} , spans from the experiment's commencement to the opening of battery 34#'s safety valve. The subsequent stage, labeled as t_{s2} , extends from the opening of 34# battery's safety valve to the opening of battery 35#'s safety valve. In the calculation process, the temperature of battery 34# is defined as the average of the three temperature measurement points. The variable ΔT represents the temperature rise difference between the two stages, while the influence of the battery's self-generated heat during this process

is disregarded. The heat transfer values from battery 34# to adjacent batteries in the two stages are presented in Table 6.

Table 6. Calorific values for each stage of the battery.

Item	t_{s1}	t_{s2}
Q_{34} (kJ)	940.5	1423.1
Q_{33} (kJ)	239.8	628.3
$Q_{33} \cdot Q_{34}^{-1}$	25.4%	44.1%
Q_{32} (kJ)	65.4	178.2
$Q_{32} \cdot Q_{33}^{-1}$	27.2%	26.4%
Q_{31} (kJ)	18.7	47.8
$Q_{31} \cdot Q_{32}^{-1}$	28.5%	26.8%
Q_{35} (kJ)	235.4	198.5
$Q_{35} \cdot Q_{34}^{-1}$	25.1%	13.9%
Q_{36} (kJ)	59.9	45.8
$Q_{36} \cdot Q_{35}^{-1}$	25.4%	23.1%
Q_{37} (kJ)	17.2	10.7
$Q_{37} \cdot Q_{36}^{-1}$	28.7%	23.4%
Q_{19} (kJ)	17.6	29.8
$Q_{19} \cdot Q_{34}^{-1}$	1.8%	2.1%
Q_{45} (kJ)	18.3	32.3
$Q_{45} \cdot Q_{34}^{-1}$	1.9%	2.2%

Table 6 illustrates that the heat generation in battery 34# is attributed to bilateral heaters and internal TR. The heat values for t_{s1} and t_{s2} are 940.5 kJ and 1423.1 kJ, respectively, with t_{s1} representing only 66.1% of t_{s2} . Here, Q_i represents the heat of a specific battery; $Q_i \cdot Q_{i+1}^{-1}$ and $Q_i \cdot Q_{i-1}^{-1}$ denote the heat ratio between adjacent batteries within a sub-module; $Q_{19} \cdot Q_{34}^{-1}$ and $Q_{45} \cdot Q_{34}^{-1}$ indicate the heat ratio between batteries across sub-modules relative to battery 34#, I denotes the cell sequence number.

Within the same sub-module, heat is conducted from the center of battery 34# to its sides predominantly through heat conduction. The presence of thermal insulation materials like aerogel affects the heat transfer fraction between adjacent batteries in t_{s1} , ranging from 25.1% to 28.5%, with an average of 26.7%. In t_{s2} , the heat transfer fraction of adjacent batteries, such as batteries 19# and 34#, differs significantly due to structural issues. The remaining adjacent batteries exhibit a heat transfer fraction ranging from 23.4% to 26.8%, averaging at 24.9%, resulting in an overall average heat transfer efficiency of 25.8%. Between sub-modules, heat is transferred from the central 34# battery to neighboring batteries primarily through radiation and convective. The heat transfer fraction via radiation and convection is relatively low due to the distance between sub-modules, averaging at only 2.0%, significantly lower than the heat conduction efficiency within the sub-module.

3.2.4. Smoke Release

Smoke emission serves as a primary TR indicator. Furthermore, because the smoke contains various combustible gases, it poses a deflagration hazard. The SRR quantifies the pack's smoke emission characteristics, as depicted by Equation (3) and illustrated in Figure 10 for the experimental duration.

$$SRR = 2.303 \frac{V}{D} \log_{10} \left(\frac{I_0}{I} \right) \quad (3)$$

where SRR is the exhaust smoke release rate, V is the exhaust unit volume flow rate, D is the exhaust shut-off diameter, I_0 is the background (pre-experiment) transmitted light intensity signal, and I is the transmitted light intensity signal during the experiment.

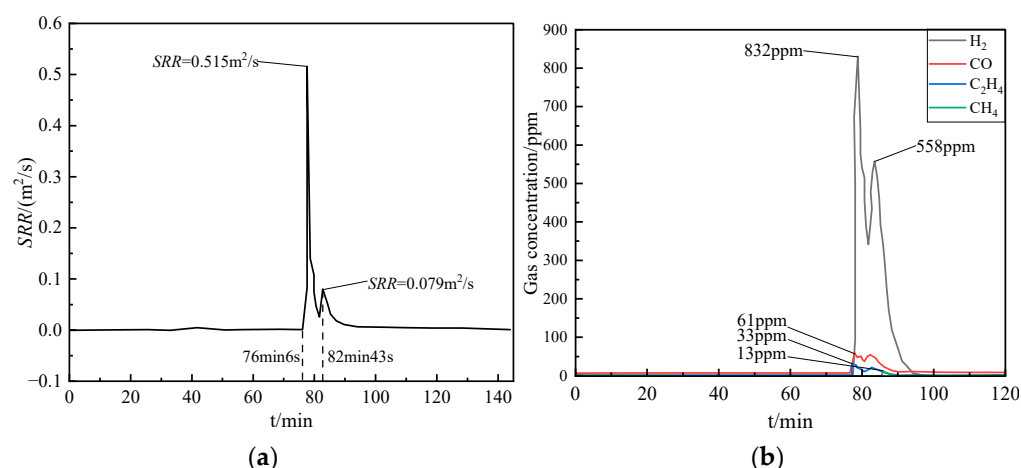


Figure 10. Smoke release rate and gas component concentration: (a) change curve of smoke release rate; (b) change curve of gas component concentration.

As illustrated in Figure 10a, the flue gas release rate during the TR process of the module began to rise significantly after 76 min 6 s, reaching a maximum value of $0.515 \text{ m}^2/\text{s}$. Battery 34# safety valve opened at 47 min 54 s. Subsequently, the flue gas gradually permeated the entire interior of the module, resulting in the expansion of the upper cover. This observation indicates that the exhaust from a single battery takes 19 min 12 s to fill the entire internal space of the module. A second peak in the flue gas release rate occurred at 82 min 43 s, with a maximum value of $0.079 \text{ m}^2/\text{s}$, which constituted only 14.3% of the first peak. The safety valve of battery 33# opened at 82 min 27 s, closely aligning with the timing of the second peak in the flue gas release rate. Throughout the smoke exhaust process, the smoke must fill the entire internal space of the module and increase the pressure to the safety valve's opening threshold before dispersing outside the module. This delay serves as an early warning signal for the TR of the pack. The flue gas is accompanied by various mixed gases, primarily consisting of H_2 , CO, CH_4 , and C_2H_4 . Figure 10b illustrates the concentration changes in each component gas throughout the gas diffusion process. The data indicate that the concentration of H_2 significantly exceeds that of the other three gases. The initial peak concentration is 832 ppm, while the subsequent peak concentration is 558 ppm. These results suggest that, when utilizing gas concentration within the cabinet as an early warning signal, the hydrogen concentration signal is the most reliable indicator.

3.3. TR Failure Mechanism

The thermal runaway (TR) failure mechanism for LiFePO_4 (LFP) single batteries and packs occurs under conditions of bilateral overheating, characterized by an initial surface temperature rise rate of $4\text{--}7 \text{ }^\circ\text{C}/\text{min}$ at a state of charge (SOC) of 100%. The primary objective is to predict the onset of TR by monitoring external physical parameters during operation, including surface temperature, temperature rise rate, voltage, exhaust behavior, and smoke release. This predictive approach aims to facilitate the termination of the TR process or its transmission.

The failure mechanism of TR in an individual battery is influenced by multiple temperature measurement points on the battery surface, the terminal voltage, and the observed exhaust behavior. Under typical conditions, the temperature should rise steadily from the combined impact of Joule heating caused by the battery's internal resistance and heat dissipation from the cooling system, while maintaining an acceptable temperature difference. When the average temperature of each battery surface exceeds a specific threshold, typically above $140 \text{ }^\circ\text{C}$, the safety valve activates. At this stage, the battery has not yet

failed and can still be interrupted. If the surface temperature reaches its maximum value or if the rate of temperature increase exceeds $1\text{ }^{\circ}\text{C/s}$, the battery experiences failure due to TR. Voltage begins to decline both before and after the safety valve opens, resulting in internal damage to the battery. As the voltage gradually decreases, the internal temperature rises until the solid electrolyte interphase (SEI) film completely collapses. This leads to direct contact between the positive and negative electrodes, causing a large-scale internal short circuit. Consequently, the voltage rapidly drops to 0, resulting in battery failure due to TR. When the battery exhibits exhaust behavior and enters the continuous gas production phase, a complete TR occurs internally, with mixed gases continuously expelled along with smoke and other solid particles. The battery ultimately fails due to TR.

The pack TR failure mechanism is assessed using multiple indicators: temperatures on the outer and battery surfaces, terminal voltage, smoke exhaust behavior, and mixed gas concentration. These conditions include a rise in the battery surface temperature inside the module reaching its maximum value or a temperature increase rate exceeding $1\text{ }^{\circ}\text{C/s}$. Additionally, a significant increase in the average temperature of the upper cover relative to the initial ambient temperature is observed, accompanied by a rapid decline in the module's total voltage. The upper cover deforms and expands, while the exhaust valve reveals the diffusion behavior of flue gas, and the gas detector registers a transient increase in hydrogen concentration.

4. Conclusions

This study conducted experimental research on 280 Ah LFP batteries experiencing thermal runaway (TR) induced by bilateral heating. The investigation encompassed both single cells and modules, analyzing the qualitative and quantitative variations in external physical parameters during TR. The primary findings are as follows.

(1) The experimental results from single battery tests indicate that the timing and temperature of TR events are random. The opening time of the safety valve varies, with a longer duration for valve activation correlating with a higher average surface temperature. This process can be categorized into four distinct stages: Initiation of TR, safety valve activation, continuous gas emission, and termination of gas emission. The proportion of hydrogen in the total gas emitted is the highest at 48.013%, making it a primary candidate for safety warning measures.

(2) The cell surface temperature exhibits three primary phenomena. First, the temperature drop phenomenon involves a brief decline in surface temperature, characterized by randomness and uncertainty, with a maximum drop rate of $1.04\text{ }^{\circ}\text{C/s}$. Second, the rapid temperature rise phenomenon is an intrinsic property of the battery, which occurs concurrently with the onset of TR. The temperature rise rate at the bottom surface of the battery is relatively stable, predominantly ranging from 1.01 to $1.87\text{ }^{\circ}\text{C/s}$. Third, the phenomenon of reaching a maximum value entails a rapid increase in temperature, culminating at a peak of $403.5\text{ }^{\circ}\text{C}$. Notably, the changes in voltage and surface temperature are largely consistent over time.

(3) The experimental results of the module: TR onset within the module is indicated earlier by the temperature signal compared to the smoke signal. Specifically, the temperature variation in the upper cover of the module, being the initial point of contact subsequent to the activation of the safety valve, serves as a visual indicator of the internal reaction progression. Heat transfer within batteries of the same sub-module primarily occurs through conduction, with an average fraction of 25.8%. Conversely, heat transfer between batteries of different sub-modules involves convection and radiation, with an average fraction of 2.0%. The release of smoke, serving as an early warning sign of TR, exhibits a hysteresis

effect, with a maximum rate of $0.515 \text{ m}^2/\text{s}$. Notably, the concentration of hydrogen within the gas mixture surpasses that of other gases, reaching a maximum of 832 ppm.

(4) The change rule of external physical quantities of the battery during the TR process—specifically, exhaust behavior, valve opening behavior, voltage and temperature changes, smoke emission behavior, and smoke release rate—indicates that when critical values are reached, the battery or module can be deemed to have failed.

In this paper, the analysis of external physical quantities involved in the TR process is limited by the experimental conditions. In practical engineering applications, TR encompasses a more complex physical field, including expansion forces and stress–strain relationships. Integrating external factors with internal battery dynamics may enhance early TR event warnings, necessitating further research.

Author Contributions: K.C.: Writing—original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. H.Z.: Writing—original draft, Visualization, Validation, Methodology, Investigation. X.W.: Writing—review and editing, Methodology, Investigation. Y.D.: Writing—review and editing, Supervision, Project administration, Investigation, Funding acquisition, Conceptualization. Y.L.: Writing—review and editing. All authors have read and agreed to the published version of the manuscript.

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