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A novel thermal runaway warning method of lithium-ion batteries

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ABSTRACT

To improve the safety of electric vehicles and battery energy storage systems, early prediction of thermal runaway (TR) is of great significance. This work proposes a novel method for early warning and short-term prediction of the TR. To give warning of TR long time in advance, a variety of battery models are established to extract key features, such as Pauta feature and Shannon entropy of voltage deviation, and then local outlier factor algorithm is used for feature fusion to detect abnormal cells. For the short-term prediction, the predefined threshold and variation rates are used. By measuring the real-time signals, such as voltage and temperature, their variation rates are calculated, based on which TR can be predicted exactly. The real data including TR from an electric vehicle are used to verify the method that it can give a warning on TR long time before it happens up to 74 days. This is remarkable for providing replacement recommendations for abnormal cells. It can also predict the occurrence of TR 33 seconds in advance to ensure the safe use of batteries.

KEYWORDS

Lithium-ion batteries, thermal runaway (TR) , early warning, local outlier factor, Shannon entropy.

Developing electric vehicles (EVs) plays an important role in building a green, low-carbon, safe and efficient energy and transport system[1]. However, the safety of batteries is constantly declining, so the risk of thermal runaway (TR) increases under extreme operating conditions such as overcharge, high temperature, collision, etc. Furthermore, battery systems often contain hundreds or thousands of cells due to the specific energy and voltage limitation of the cell. The TR of any cell in the battery system may cause the chain TR of the surrounding cells, leading to serious fire accidents with casualties and huge economic losses[2]. Therefore, developing early warning method of TR is of great significance to ensure the safe use and maintenance of battery systems.

TR is usually caused by an uncontrolled temperature rise triggered by a series of chain exothermic reactions inside the battery, which eventually evolves into fire and explosion[3]. The factors that lead to TR of the cell mainly include external factors[4] and internal factors[5]. On the one hand, external factors like collisions or extrusions[6], may damage the battery separator, leading to a violent reaction between the positive and negative materials within the cell. This can trigger a chain reaction of exothermic reactions between adjacent cells, ultimately resulting in a severe TR accident. Accidents caused by external factors, especially mechanical damage, happen suddenly and violently, so it could hardly be foreseen but easily detected. On the other hand, the internal factors are intricate and diverse, and are closely related to the dynamic working conditions and aging state of batteries due to the complex internal structure and electrochemical reactions of batteries[7]. The anode and cathode plates are not completely uniform and smooth, and the electrolyte may also contain a small amount of tiny metal debris[8]. These small defects or inconsistencies do not affect the performance in the early lifetime. However, extreme working conditions will cause lithium ions to gradually precipitate at the defect position and form lithium dendrites in long-term use[9], which will puncture the separator and induce internal short circuit[10], thus resulting in TR of the system[11]. The TR caused by internal factors often lasts for a long time, so it is possible to be predicted by analyzing the external characteristics of the cell.

The existing TR warning methods can be divided into two categories. One is measurement-based and the other is model-based[12]. Signals that can be directly measured include temperature[2], voltage, pressure[13], gas[14], ultrasonic detection[15], etc. Temperature is the most critical and universal characteristic to judge whether TR occurs and which stage of TR the battery is in. However, the heat transfer is slow in the battery pack, so the TR detection based on temperature has a long delay. In addition, when TR occurs, there are other easily detectable phenomena, such as voltage drop, battery swelling and the release of flammable gas. Such measurement-based methods are passive detection of significant changes in the TR process. They are characterized as simple and reliable, but it is hard for them to predict TR long time in advance. Model-based methods use characteristic parameters extracted from physical or statistical battery models. For example, Liu et al.[16] proposed an internal temperature estimation method, which combined a second-order resistance-capacitance equivalent circuit model (ECM) with a two-state thermal model to estimate the internal temperature of lithium ion batteries based on limited numbers of temperature sensors. This method solved the problem of serious lag in temperature propagation to a certain extent. Zhao et al.[17] established an fractional order model and applied distribution of relaxation times to calculate the internal resistance. The fractional-order extended state observer was applied to update the internal resistance. The change of the internal resistance was faster and more significant than that of the voltage, which gave it a chance to warn TR earlier in advance. Ding et al.[18] proposed meta TR forecasting neural network using voltage and temperature to perform multistep ahead forecast accurately for battery TR state at cell-level. In addition, some methods describing battery aging or damage are used for long-term TR warning, such as electrochemical impedance spectroscopy (EIS), incremental capacity analysis

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(ICA)[19], differential voltage analysis (DVA) and the relaxation time distribution, which reveal the changes of ohmic internal resistance, the internal resistance of the solid electrolyte interface (SEI) film, charge transfer resistance and solid phase diffusion resistance during the long-term evolution of TR. In recent years, there are also many deep learning–based methods are applied for early warning. Ma et al.[20] proposed a voltage-temperature joint TR warning method based on deep learning with attention mechanism, which achieved 8–13 min ahead TR prediction in real-world scenarios. Kim et al.[21] proposed a multiphysics-informed neural network to predict TR. This model was encoded with the governing laws of physics, including the energy balance equation and Arrhenius law, and thus received a high prediction accuracy.

Despite good progress in modeling and simulating TR evolution, warning the TR long time in advance remains challenging.

(1) Current methods are difficult to deal with low quality real data containing noise and interference. For example, the sampling interval is very large limited by the communication cost in the big data platform.

(2) Many novel models perform well in certain scenarios, but struggle to adapt to others. A framework that can incorporate the advantages of multiple models is urgently needed.

Therefore, this work proposes a new method for long-term warning and short-term prediction of TR. The long-term warning part is based on the local outlier factor (LOF) algorithm[22] fusing multi predicting models to warn TR long time in advance. When an abnormal battery is detected but continues to be used, it can indicate a significant risk of TR, but whether and when TR occurs is not very clear. Therefore, the predefined threshold and variation rate of key parameters are also included as the short-term prediction part to warn the impending TR. A novel framework proposed in this paper is that using LOF method to fuse different kinds of models to improve the performance of long-term warning, and using short-term prediction to ensure human escape and fire extinguishing in the impending TR. The LOF algorithm is used to integrate several statistical models for long-term early warning since the sampling interval is more than 50 seconds. However, this open architecture for early warning can integrate different types of models so it can be applied in a variety of scenarios. For example, the ECM[23] and the pseudo two-dimensional (P2D) model[24] can be added when the sampling interval is small and stable. If new signals such as electrode potential and internal pressure are measured, the mechanism model can also be extended within the framework. In short, this method provides a framework where different models can be fused to improve the performance of early warning. The rest of this paper is organized as follows. The structure, working principle and flow of early warning are elaborated in Section 1. In Section 2, the validation and discussions of the short-term prediction and long-term warning of TR are outlined. Conclusions are finally drawn in Section 3.

1 Methodology

1.1 Overview

The overview of the proposed method is shown in Figure 1. EVs upload real-time battery operating data to the National Monitoring and Management Platform of Electric Vehicles (NMMP-EVs), where the warning method of TR runs to provide early warning advice for EVs.

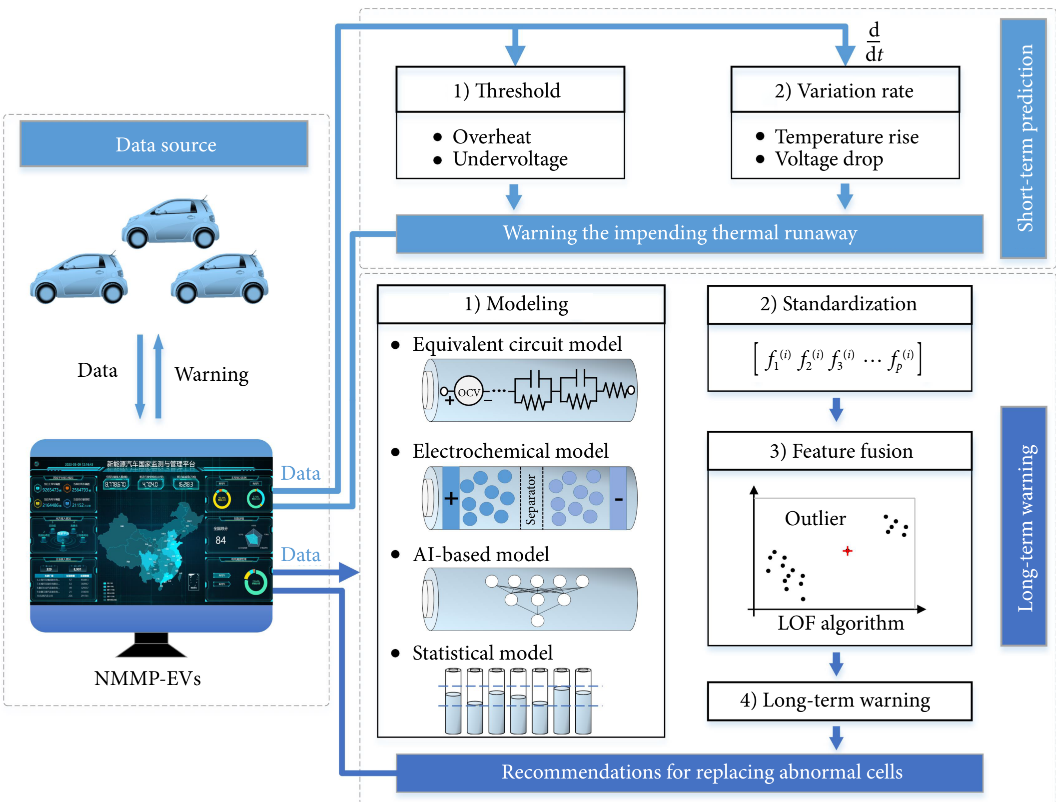

Fig. 1 Method overview.

In the very early stage of TR evolution, the voltages of normal and faulty cells are so similar that they are difficult to distinguish.

To enlarge this tiny difference, several battery models are applied to extract the characteristic parameters. After that, these parameters of cells are standardized and shaped into vectors. Then the shaped parameters are fused by the LOF algorithm to look for the outlier cell. Finally, the outlier cell is found and the TR is warned long time in advance. The method for long-term warning of TR provides a general framework for fusing TR features, whether from mechanical or statistical or other models. For example, the capacity and internal resistance can be calculated using the equivalent circuit model, while certain dangerous side reactions can be monitored through a P2D model. Moreover, valuable TR features can be obtained from statistical or machine learning models. All features can be used as input to the LOF algorithm, and the fusion of features can be realized to improve the warning performance. However, the average sampling interval of TR data used in this paper is 50 seconds, which makes the dynamic modeling hardly practical[25]. In this paper, several models based on voltage inconsistency are applied to overcome this large sampling interval, such as the Pauta criterion model and the Shannon entropy model.

With this method, TR will be warned long time in advance. However, it is unclear whether and when TR occurs. The predefined threshold and variation rates of key parameters are also applied to predict the impending TR. It predicts the onset of TR by analyzing sensor data, including voltage, current, temperature and their rate of change. This method for short-term warning of TR runs in real time, so it can be quick and reliable for warning the impending TR.

1.2 Pauta criterion model

The Pauta criterion, also known as the three sigma criterion[26], assumes that the signal follows a normal distribution. A sampling point that deviates from the mean by more than 3 times the standard deviation is considered as impossible (a probability of 0.0026), i.e., an anomaly, as shown in Figure 2(a). The Pauta criterion gives an early warning by comparing whether the sampling point deviates from the mean by more than 3 times of the standard deviation. In fact, due to the small number of cells and the difference between the actual distribution of voltages and the normal distribution, the cell voltages are often beyond the 3σ range due to differences in working conditions, temperature and other factors. Therefore, defining the ratio of absolute deviation to standard deviation and determining the appropriate threshold according to the actual situation can effectively avoid misdiagnosis. In addition, the Pauta criterion has great fluctuations, which is difficult to be applied in practice when dealing with single frame of sampling data. Calculating the average value of Pauta feature over a period of time can suppress the fluctuation and improve the stability of the Pauta feature. Thus, an early warning feature based on the Pauta criterion is defined as follows

$$F_1(i) = \frac{1}{W}\sum_{w=1}^{W}\frac{|e_{i,w}|}{\sigma_w} \tag{1}$$

where, $w = 1, 2, \ldots, W$, W indicates the length of the sampling window. $i = 0, 1, \ldots, N$, N indicates the number of cells. $e_{i,w}$ and σ_w are the voltage deviation of the i-th cell and the standard deviation of all cells at the time step w, respectively, whose definitions are as follows

$$e_{i,w} = x_{i,w} - \bar{x}_w \tag{2}$$

$$\bar{x}_w = \frac{1}{N}\sum_{i=1}^{N} x_{i,w} \tag{3}$$

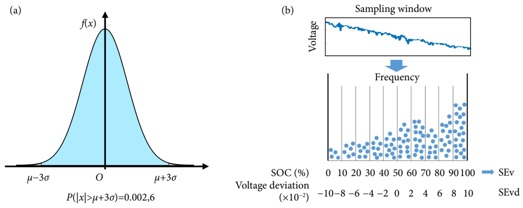

Fig. 2 Pauta criterion and Shannon entropy. (a) Pauta criterion and **(b)** Shannon entropy.

$$\sigma_w = \sqrt{\frac{1}{N}\sum_{i=1}^{N} x_{i,w}^2} \tag{4}$$

where, x_i and $\bar{x}_w$ are the voltage of the i-th cell and the average voltage of all cells at time step w, respectively.

1.3 Shannon entropy model

Shannon entropy is a measure of how much information data contains. The Shannon entropy increases with the increase of data uncertainty, which is defined as

$$SE = -\sum_{m=1}^{M} p_m \log_2(p_m) \tag{5}$$

where SE is the Shannon entropy, M is the number of intervals (we set M to 10), and p_m is the frequency at which the sampling point falls within the m-th interval.

The Shannon entropy of voltages (SEvs) use 11 voltage points to divide the voltage range 2.5 V–3.65 V into 10 unequal small intervals. These 11 voltage points are obtained when the state of charge (SOC) reaches 0, 10%, …, 100% in the small constant current charging test. In the Shannon entropy of voltage deviations (SEvds), the voltage deviation range −0.1 V–0.1 V is divided into 10 equal intervals, as shown in Figure 2(b).

1.4 Local outlier factor model

Different warning features are sensitive to specific types of accidents, but not to other types of accidents, which is prone to missing and false alarms. Therefore, the warning method based on a single feature has limitations. The Pauta feature, Shannon entropy of voltage deviation, mean and standard deviation of cell voltage are fused by LOF method for accurate long-term warning.

The LOF model describes the outlier characteristics of a cell according to the distance distribution of each cell feature vector in a module, and screens the outlier cell with its abnormal behavior. All cells are connected in series to form battery modules so the current flowing through them is same. However, the voltage behaviors of each cell are different, and can be used for anomaly detection. For the battery module with parallel parts, its parallel parts can be regarded as a big cell in the LOF model. The detection result can be applied for further detection with the current and internal resistance to distinguish the anomaly in the parallel parts.

In this model, each cell is represented by a feature vector, which includes the Pauta feature, the Shannon entropy, the average voltage as well as the standard deviation of voltage within the sampling window. The feature vector set composed of each cell in the module is set to $\boldsymbol{S}$. For any vector $\boldsymbol{p}$ and vector $\boldsymbol{o}$ in $\boldsymbol{S}$, $d(\boldsymbol{p}, \boldsymbol{o})$ is defined as the Euclidean distance between vector $\boldsymbol{o}$ and vector $\boldsymbol{p}$.

The distances between the vector $\boldsymbol{p}$ and any vector in the set $\boldsymbol{S}$ are calculated, sorted in ascending order and labeled the vectors as $\boldsymbol{o}_1, \boldsymbol{o}_2, \ldots, \boldsymbol{o}_{N-1}$. N is the total number of vectors in the set $\boldsymbol{S}$, which is the number of cells in the module. Then there is

$$d(\boldsymbol{p},\boldsymbol{o}_1) \leqslant d(\boldsymbol{p},\boldsymbol{o}_2) \leqslant \cdots \leqslant d(\boldsymbol{p},\boldsymbol{o}_{N-1}) \tag{6}$$

$d_m(\boldsymbol{p})$ is defined as the m-distance of the vector $\boldsymbol{p}$, i.e., the distance from the vector $\boldsymbol{p}$to its m-th nearest vector, where

$$d_m(\boldsymbol{p}) = d(\boldsymbol{p},\boldsymbol{o}_m) \tag{7}$$

The m-neighborhood of the vector $\boldsymbol{p}$ is defined as the set of vectors, whose distance from vector $\boldsymbol{p}$ is no greater than m-distance of $\boldsymbol{p}$. It is denoted as

$$N_m(\boldsymbol{p}) = \{\boldsymbol{o} | d(\boldsymbol{p},\boldsymbol{o}) \leqslant d_m(\boldsymbol{p}), \boldsymbol{o} \in \boldsymbol{S}\} \tag{8}$$

The reachable distance of vector $\boldsymbol{p}$ with respect to vector $\boldsymbol{o}$ is defined as

$$rd(\boldsymbol{p},\boldsymbol{o}) = \max\{d_m(\boldsymbol{o}), d(\boldsymbol{p},\boldsymbol{o})\} \tag{9}$$

Apparently,$d(\boldsymbol{p},\boldsymbol{o}) = d(\boldsymbol{o},\boldsymbol{p})$, but $rd(\boldsymbol{p},\boldsymbol{o}) \neq rd(\boldsymbol{o},\boldsymbol{p})$.

In the set $\boldsymbol{S}$, to calculate the density of the points around the vector $\boldsymbol{p}$, the locally reachable density of vector $\boldsymbol{p}$ is defined as

$$lrd(\boldsymbol{p}) = 1/\left(\frac{\sum_{\boldsymbol{o}\in N_m(\boldsymbol{p})} rd(\boldsymbol{p},\boldsymbol{o})}{|N_m(\boldsymbol{p})|}\right) \tag{10}$$

where $|N_m(\boldsymbol{p})|$ is the number of vectors in the m-neighborhood $N_m(\boldsymbol{p})$.

The smaller locally reachable density of vector $\boldsymbol{p}$ means the vectors in its m-neighborhood are more spread out. According to this idea, the local outlier factor $LOF(\boldsymbol{p})$ is defined as follows

$$LOF(\boldsymbol{p}) = \frac{1}{|N_m(\boldsymbol{p})|}\sum_{\boldsymbol{o}\in N_m(\boldsymbol{p})}\frac{lrd(\boldsymbol{o})}{lrd(\boldsymbol{p})} \tag{11}$$

1.5 Early warning flow

The calculation and warning flow of the proposed method is shown in Table 1.

Table 1 calculation algorithm

Steps	Details
1. Initialization	Set the size of the sampling window Set k = 1
2. Features calculation	Get the voltage data in the sampling window Calculate the Pauta feature, the Shannon entropy of voltage deviation, the mean and standard deviation of the cell voltage
3. *LOF* calculation	Calculate *LOF*
4. Threshold detection	If *LOF* is greater than its warning threshold, go to step 5 If not, set k = k+1, go to step 2
5. Long-term warning	Warn the TR

2 Results and discussion

2.1 TR dataset

Data from an EV experiencing a TR accident is used for validation, which includes complete usage data for the three months before the TR accident. This accident data is derived from the National Monitoring and Management Platform of EVs (NMMP-EVs)[27] and the average sampling interval is 50 seconds. There is no abuse such as overcharging and overdischarging according to the data recorded by the NMMP-EVs, the specific cause of the TR is unknown. However, it is confirmed that the TR first appears in cell #85 based on the alarm signal and temperature. This EV travels a total of 36,980 kilometers before TR. The pack voltage, pack current, SOC and the maximum and minimum voltages of the battery system are shown in Figure 3. The battery pack is a lithium-ion battery pack with $LiNi_xMn_yCo_{1-x-y}O_2$ (NMC) as the positive electrode material and graphite as the negative electrode material. It includes 96 cells in series. The average daily electricity consumption of this EV in the three months during this period is 152.92 Ah, equivalent to about 101.95% of the nominal capacity.

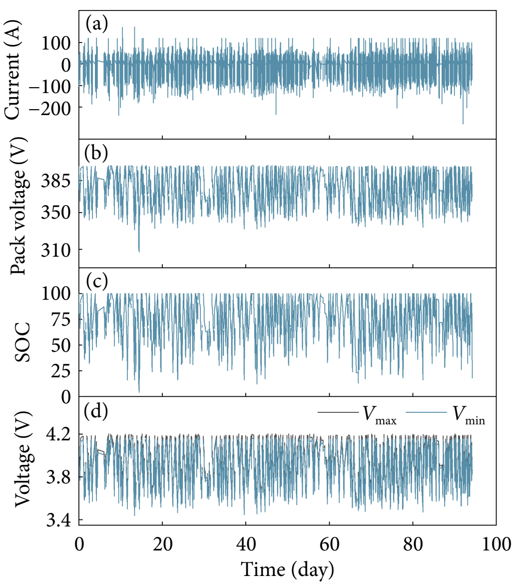

Fig. 3 Battery pack usage data for 3 months before TR. (a) The pack current, **(b)** the pack voltage, **(c)** the SOC and **(d)** the maximum and minimum voltages of cells in the pack.

2.2 The short-term prediction of TR

The measurement-based method for short-term prediction of TR can accurately predict the exact time when TR is about to occur actually with the predefined threshold and variation rates from key parameters as shown in Figure 4. The normal operating temperature of the battery is lower than 55 °C. When the temperature exceeds 55 °C, the TR is predicted and may evolve into a fire accident within a few minutes. The threshold of temperature-based warning is set as 55°C. The temperature-based warning is a very common method, so it is set as the reference point of the time axis to show the performance of variation rates-based warning in Figure 4. When the voltage drop rate exceeds 0.1V/s and lasts for 3 seconds, the TR is predicted 12 seconds ahead of the temperature-based warning. When the temperature rise rate exceeds 2 °C/s, the TR is predicted 7 seconds ahead of the temperature-based warning. Many signals and their variation rates can be used to reduce false alarm.

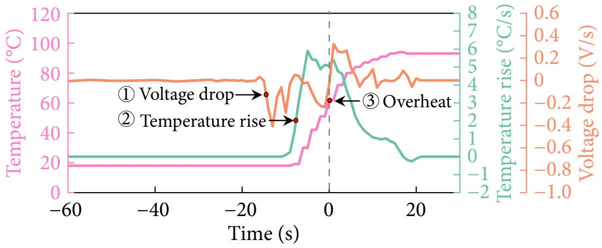

Fig. 4 Results of the short-term prediction of TR.

2.3 The long-term warning of TR

In the method for the long-term warning of TR, many key features are extracted, standardized and fused in the LOF algorithm to predict the TR long time in advance. Their performance is as follows.

2.3.1 Pauta criterion

Results of Pauta feature are shown in Figure 5. It shows the cell #47 with the highest F_1 value, #92 with the lowest F_1 value and #85 with TR. The Pauta feature of most cells is similar to that of normal cell #92. Although the Pauta feature well highlights the differences between different cells, all cells do not exceed the warning threshold even when the TR occurs. This phenomenon may be due to the excessive internal resistance of cell #47, or the difference between the SOC of cell #47 and other cells. This reveals that the imbalance of the battery pack may cause Pauta feature to make false alarms, so a better way to use this feature is needed.

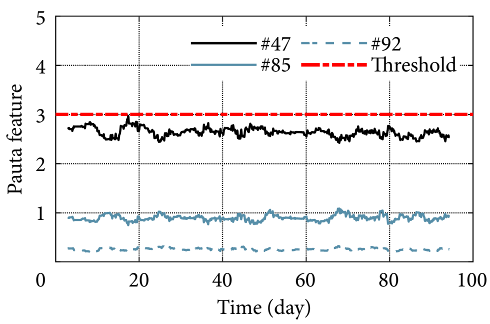

Fig. 5 Warning with Pauta feature.

2.3.2 Shannon entropy of voltage deviation

The input of *SEvd* is the deviation of each cell relative to the average voltage, so the voltage fluctuations caused by drastic current changes can be well suppressed. Compared with the *SEv*, the *SEvd* can amplify the voltage inconsistency between cells, and restrain the fluctuations caused by the working conditions, so as to better warn the accident. Figure 6 shows the *SEv* and *SEvd* curves of two typical cells. #85 is an abnormal cell, and #21 is a normal cell. The *SEv* is greatly affected by the working condition and fluctuates sharply. The difference between the curves of cell #85 and cell #21 is not obvious. Although the *SEvd* of cells #85 and #21 also fluctuate a little, the difference between them is significant, indicating that *SEvd* is better than that of *SEv* in diagnosing abnormal cells. Actually, the voltage is strongly affected by the current due to battery polarization, and thus SEv fluctuates wildly with the conditions. SEvd uses the voltage deviation instead of the voltage to calculate the Shannon entropy, which enlarges the difference between cells and restrains the voltage fluctuation caused by the working condition at the same time.

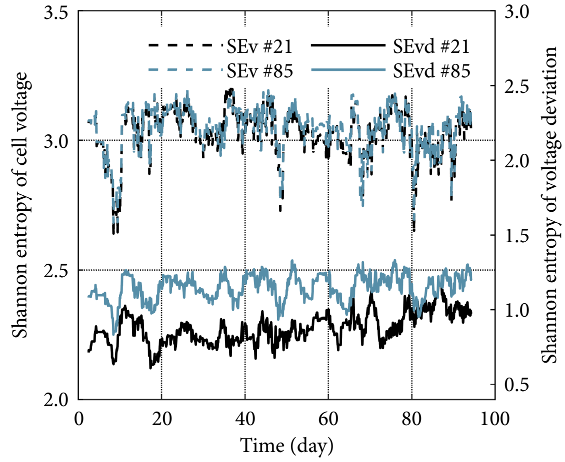

Fig. 6 Shannon entropy of cell voltage and Shannon entropy of voltage deviation.

2.3.3 Local outlier factor

Although the Shannon entropy of voltage deviation can be used to detect the abnormal cell intuitively, it is still difficult to meet the requirement of automatic identification of abnormal cell through threshold detection. In order to further exaggerate the difference between abnormal cells and normal cells and keep the threshold stable, the LOF algorithm is used to further deal with these warning features. This method uses the Pauta feature, Shannon entropy of voltage deviation, mean and standard deviation of cell voltage to characterize each cell, calculates the distance of each cell in the characteristic space, and uses the LOF to evaluate the dissimilarity of each cell. Usually, the cells in a module have similar physical and chemical properties and working conditions, so the *LOF* is low, usually around 1. When there is an abnormal cell, its high *LOF* is easy to detect by setting an early warning threshold. This makes the threshold of LOF stable, so it is easy to detect abnormal cells by threshold detection. Results of the LOF method are shown in Figure 7. The warning threshold is set to 2 so that the other cells don't exceed the threshold all the time. The *LOF* of cell #85 exceeds the warning threshold on the day 20, so this method warns TR 74 days in advance. In addition, the *LOF* of cell #85 shows an upward trend in the three months before TR, and exceeds the warning threshold several times, and finally TR occurs. In contrast, the *LOF* of normal cell #21 basically remains fluctuating around 1. This indicates the multi predicted models co-driven method for long-term warning has good performance in avoiding the TR.

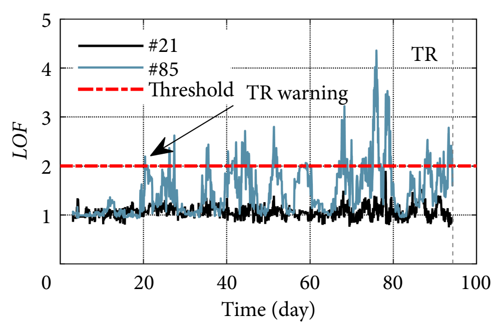

Fig. 7 Warning results of LOF method.

2.3.4 The sampling window size

The sampling window size determines the amount of information to be input and processed, which has an impact on the long-term

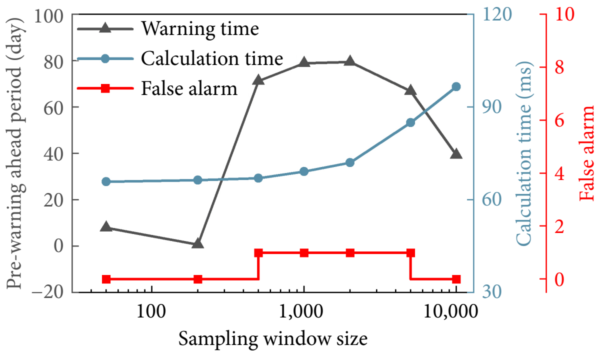

Fig. 8 Performance with different sampling window sizes.

warning performance and calculation cost. Since the TR data is sampled at uneven intervals, the sampling window size is defined here in terms of the number of sampling points. The alarm is triggered only when the threshold is exceeded for three consecutive times. At the same time, because the sampling window size will affect the range of *LOF* and thus the early warning threshold, it is impossible to use a unified threshold to compare the performance under different sampling window sizes. Therefore, the performance of the algorithm is evaluated by the maximum pre-warning ahead period without causing normal battery false alarms. Figure 8 shows the maximum pre-warning ahead period, single calculation time cost and false alarm under different sampling window sizes. The horizontal coordinate is the sampling window size, using the logarithmic coordinate axis. The single calculation time cost is defined as the time taken to calculate the *LOF* of each cell in a sampling window. The device used in this paper is HP Z820, the processor is dual-core Intel(R) Xeon(R) CPU E5-2643 v2@ 3.50 GHz, and the memory is 64.0 GB. The calculation is performed using MATLAB R2019a version.

With the increase of the sampling window size, the maximum pre-warning ahead period firstly increases and then decreases, and has the maximum at 2000. With the increase of the sampling window size, the amount of information increases, so the single calculation time cost gradually increases. When the sampling window size is less than 2000, the single calculation time is almost the same, showing a slow increase trend. When the sampling window size is larger than 2000, the single calculation time increases rapidly. The statistics of false alarm show that there are false alarms when the sampling window size is less than 500 or greater than 10000. Therefore, the optimal sampling window size is 2000, which contains an average of 27.8 hours of data.

In short, the Pauta feature, Shannon entropy of voltage deviation, mean and standard deviation of cell voltage are applied to characterize each cell in the long-term warning method, and the LOF algorithm is then used to identify the abnormal cell and warn the TR. This method warns TR caused by cell #85 74 days in advance, which helps to replace the abnormal cell and avoid the severe TR accident.

3 Conclusions

This work proposes a novel method for early warning and short-term prediction of TR , which effectiveness is verified by data from an EV with the TR accident. The main conclusions are as follows:

(1) For early warning, it is proposed to use Shannon entropy of voltage deviation to characterize battery inconsistencies, which reduces the influence of operating conditions and significantly improves the characterization ability compared with Shannon entropy of cell voltage. By using real data, it is found that the proposed method can predict TR 74 days in advance.

(2) For short term prediction, the threshold and variation rates of key parameters from different sensors are used to obtain the exact time for the occurrence of the impending TR, which helps to extinguish fire and evacuate people in time to avoid serious fire accidents.

This method provides an open framework for the collaboration of different types of models to obtain a better warning performance and adapt to more usage scenarios. It will help detect abnormal cells and replace them in time to improve the safety of the battery system and avoid serious thermal runaway accidents.

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Additional information

Declaration of competing interest

The authors have no competing interests to declare that are relevant to the content of this article.

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