

Article

Coupling Model and Early-Stage Internal Short Circuits Fault Diagnosis for Gel Electrolyte Lithium-Ion Batteries

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Abstract

This paper presents a method for modeling and predicting ISC in gel-electrolyte lithium-ion batteries, addressing critical safety concerns in electric vehicles. While gel-electrolytes are highlighted for their superior stability and performance advantages over liquid-electrolytes, they remain susceptible to IISC due to factors such as dendrite formation or mechanical stress. This study provides a detailed analysis of the unique ISCs mechanism in gel-electrolytes, emphasizing the differences between gel-electrolyte and liquid-electrolyte batteries in terms of ion transport dynamics and thermal performance. Based on these characteristics, an electrochemical–thermal–ISC coupling model was developed, and an external short-circuit resistance test was conducted to validate the model’s accuracy. By simulating various ISC states using the coupling model, a comprehensive dataset of battery ISC parameters was obtained, encompassing voltage, current, temperature, SOC, capacity loss, and internal resistance. ISC prediction models were subsequently developed using BP, CNN, and LSTM networks, with a comparative analysis of their prediction accuracy. This research advances the ISC prediction framework for gel-electrolyte batteries and demonstrates the potential of CNN-based models to achieve higher accuracy in fault prediction. Accurate ISC prediction is crucial for ensuring safe battery operation in electric vehicles.

Keywords: gel-electrolyte; lithium-ion batteries; internal short circuits; ISC prediction model; fault prediction



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1. Introduction

Lithium-ion battery is a complex electrochemical system. In recent years, lithium-ion battery failures due to internal short circuits (ISCs) have been a significant concern to the entire lithium-ion cell market from consumer electronics to electric vehicles. Currently, liquid-electrolyte lithium-ion batteries have characteristics of easy leakage, flammability, and volatility, all potential safety hazards [1,2]. Therefore, safer and more efficient gel-electrolyte lithium-ion batteries have become a hot research topic for researchers [3,4]. Gel-electrolyte lithium-ion batteries provide several advantages, including high energy density, long cycle life [5,6], excellent discharge rates, and resistance to leakage [7,8] and to the formation of lithium dendrites [9,10]. Furthermore, they exhibit superior ionic conductivity and enhanced scalability [11–13]. Consequently, gel-electrolyte lithium-ion batteries may be susceptible to internal short-circuit (ISC) failures, a phenomenon also observed in liquid-electrolyte-based systems [14,15].

The mechanism of battery ISCs is highly complex, and current ISC diagnosis methods primarily rely on model-based and data-driven approaches. Model-based diagnostic methods identify ISCs by comparing the difference between a lithium-ion battery model and real-world data against a predefined threshold [16,17]. Kong X. et al. [18] conducted simulation experiments using a pseudo two-dimensional model and an impedance identification method, revealing that the effective conductivity of the separator is strongly correlated with the degree of internal short-circuiting in batteries. Sheikh M. et al. [19] performed mechanical failure analysis of thermal runaway propagation in 18650 lithium-ion batteries by developing a battery model through finite element analysis and numerical simulation techniques. Their experiments validated the accuracy of the numerical simulation model and demonstrated its utility in predicting the initial failure of batteries. Feng X. et al. [20] proposed a model-based online ISC diagnosis algorithm that utilizes the average voltage value of the battery pack. The algorithm detects ISC faults by calculating the deviation of the evaluated voltage and incorporates temperature information to enhance the robustness of the diagnosis process.

Data-driven diagnostic methods are based on a large amount of historical data on ISCs in lithium-ion batteries, extracting battery fault characteristics and using machine learning or deep learning algorithms for training. This method does not require electrochemical modeling of lithium batteries, which is relatively simple and accurate. Ojo O. et al. [21] proposed a data-driven thermal fault detection method based on the estimation of battery temperature with Long and Short Term Memory (LSTM). Samanta A. et al. [22] made a review and analysis of machine learning algorithms on lithium-ion battery safety diagnosis for the three learning methods categorized as supervised learning, unsupervised learning and reinforcement learning; the most widely used is based on the supervised learning methods, such as artificial neural network (ANN), random forest (RF), support vector machine (SVM), among which ANN algorithm is the most commonly used.

Challenges and problems:

- (1) Because of the difference in ion transport and thermal performance between gel-electrolyte battery and liquid battery, there is still a gap in the research of internal short circuit safety of gel-state lithium-ion batteries.
- (2) The traditional methods require the establishment of modified pseudo two-dimensional models, three-dimensional electrochemical thermal internal short circuit coupling models, or equivalent circuit models. Algorithms also require complex methods such as recursive least squares, which have complex models, large computational complexity, and high practical application thresholds.
- (3) The traditional method is relatively simple, relying on threshold values and other judgment criteria. However, in actual situations, determining the parameter threshold is challenging as it requires consideration of multiple factors, resulting in low accuracy. Meanwhile, the diagnostic method based on ISC mechanisms and models involves complicated calculations, making it difficult to apply in practice.
- (4) Data-driven diagnostic methods require extensive ISC data, but conducting a large number of battery ISC experiments poses certain safety risks, and these experiments exhibit poor repeatability and controllability.

Contributions of this work:

The safety problem of liquid lithium-ion batteries due to internal short circuit is the focus of current research. Traditional ISC diagnosis method has the problems of complex calculation, low accuracy and difficult data acquisition. This paper solves the problem of controllable recurrence in ISC tests by combining model simulation and experimental testing. The ISC method proposed in this paper demonstrates both simplicity and accuracy, with great application potential.

- (1) This study focuses on analyzing the thermal runaway process and mechanism of ISC in gel-electrolyte lithium-ion batteries. It provides a detailed analysis of the unique ISC mechanism in gel-electrolytes, highlighting the differences between gel-electrolyte batteries and liquid-electrolyte batteries in terms of ion transport and thermal performance. The research results can provide theoretical support for the safety design of battery cells and systems.
- (2) A battery electrochemical–thermal–ISC coupling model for lithium-ion batteries is established, which effectively addresses the challenge of acquiring ISC data by replacing physical ISC tests with simulation methods. Through the combination of model simulation and experimental testing, this approach solves the problem of poor controllability and repeatability in internal short circuit testing.
- (3) ISC prediction model grounded in deep learning was established. Deep learning model with both simplicity and accuracy and great application potential is established. Reduce complexity and improve accuracy through optimizer selection, learning rate strategy and architecture optimization.

The safety issue of liquid lithium-ion batteries arising from ISCs is a critical focus of current research. Traditional ISC diagnosis methods suffer from limitations such as complex calculations, low accuracy, and challenging data acquisition. This paper addresses these challenges by integrating model simulation with experimental testing, thereby solving the problem of controllable recurrence in ISC testing. The proposed ISC method demonstrates simplicity, high accuracy, and significant application potential.

Organization of the paper:

Section 2 presents the methodology analysis, Section 3 focuses on the mechanistic analysis of ISCs and battery ISC model designs, Section 4 details the model validation and dataset acquisition process, and Section 5 outlines the methods for predicting ISCs. The key conclusions are summarized in Sections 6 and 7. The Structure of research technology is shown in Figure 1.

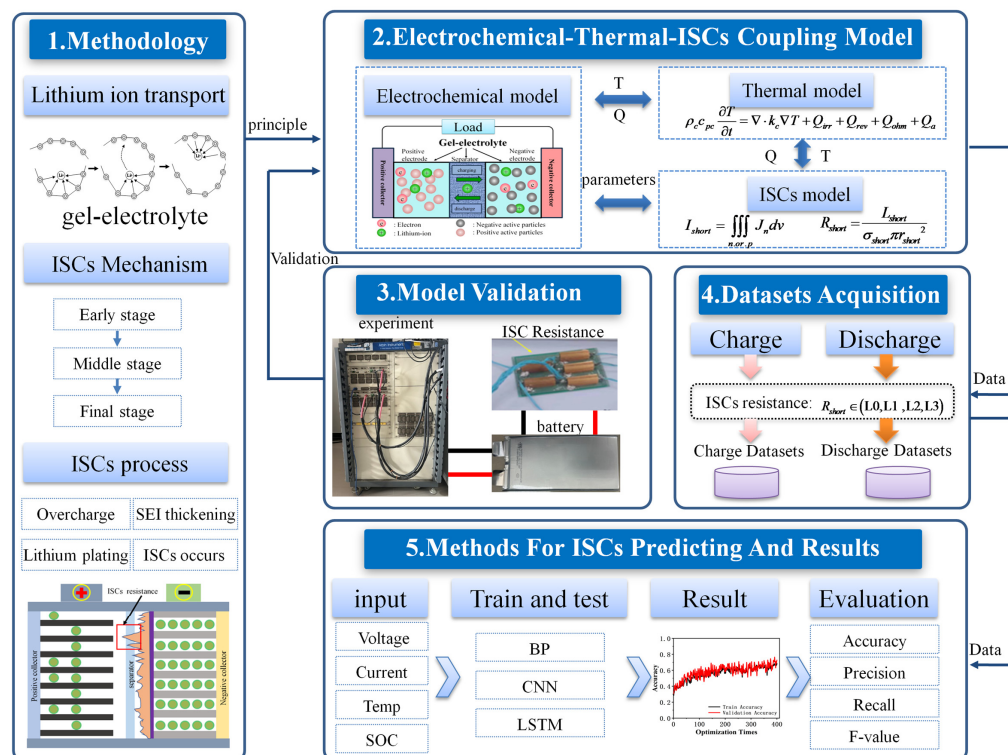


Figure 1. Structure of research technology.

2. Methodology

2.1. Principle

The structure of gel-electrolyte lithium-ion batteries consists of a negative electrode material, a positive electrode material, and a separator, as shown in Figure 2.

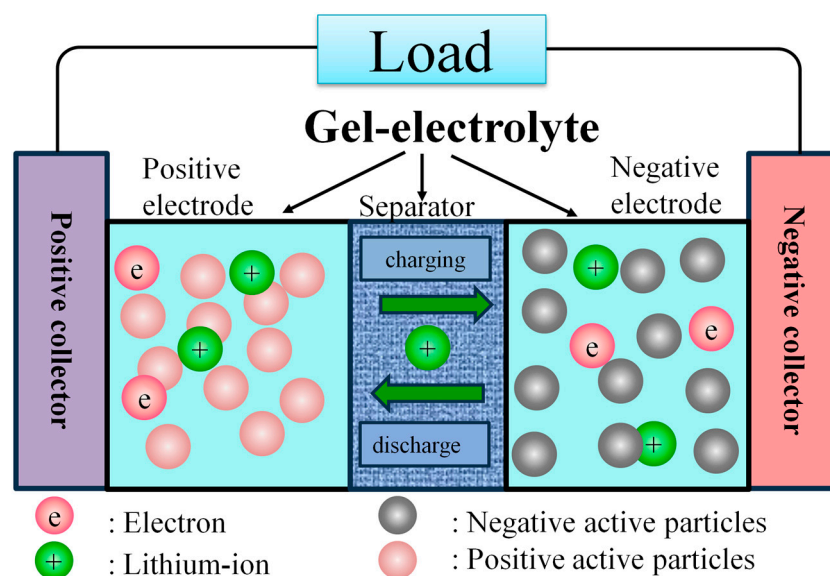


Figure 2. Structure of gel-electrolyte lithium-ion batteries.

Gel-electrolytes are composed of a polymer matrix enriched with a solvent and lithium salt. The solvent plays a crucial role in dissolving ions, which enables the transport of lithium ions through the electrolyte as they migrate along the polymer chains. This sequential movement facilitates the efficient directional transport of lithium ions, as shown in Figure 3. The gel polymer electrolyte consists of a complex microporous network formed by a polymer matrix, plasticizers, and compatible lithium salts. The solvent enables ion dissolution, allowing lithium ions to migrate through the electrolyte as the polymer chains move, ultimately facilitating the directional transport of lithium ions. The lithium salts within the electrolyte are responsible for conductivity, while the polymer provides mechanical strength to the electrolyte. Additionally, the electrolyte can act as a plasticizer within the polymer matrix, lowering the glass transition temperature and thereby enhancing the ionic conductivity of the battery. The safety of gel-electrolytes is influenced by the degree of fluid retention in the polymer. At the interface with the electrodes, the electrochemical stability of liquid-electrolytes is significantly lower than that of solid polymer electrolytes, resulting in a lower decomposition potential for gel-electrolytes.

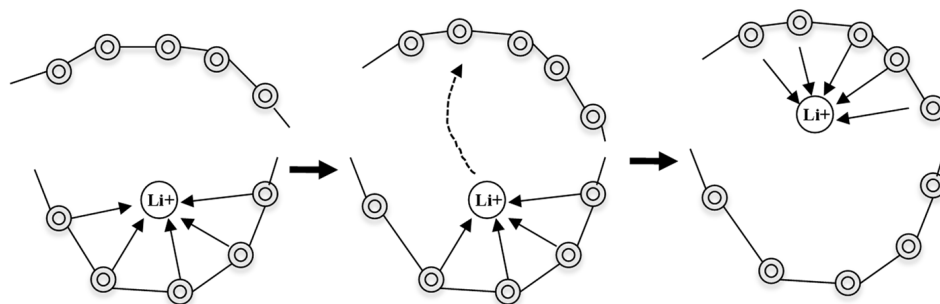


Figure 3. Lithium-ion transport in the gel-electrolyte.

2.2. Mechanism Analysis of ISCs

Under complex and severe conditions such as mechanical, electrical, and thermal abuse, gel-electrolyte lithium-ion batteries may suffer ISCs [23]. As the internal temperature of a lithium-ion battery rises, it can trigger a variety of reactions among the electrode materials, electrolyte, and gel matrix. This phenomenon serves as an early warning sign of potential ISCs. Should the temperature continue to escalate, it may further exacerbate the development of these ISCs. Such short circuits can occur at any point during the battery's operational cycle. Based on the observable characteristics of the battery's behavior, ISCs can be classified into early stage, middle stage, and final stage [24,25], as shown in Figure 4. Different stages of ISCs have significant differences in thermal and electrical characteristics, and the development speed of ISCs varies. The characteristics of the three stages are mainly as follows:

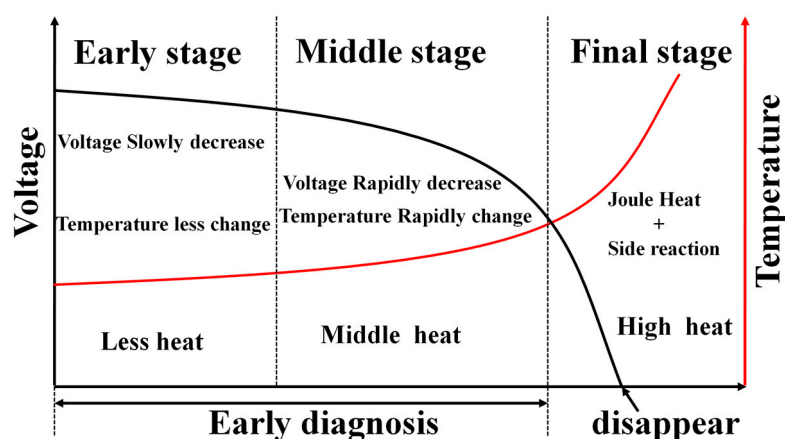


Figure 4. Battery ISC stage.

Early stage: The resistance of the internal short-circuit resistor is high, the voltage drop of the battery is slow, the Joule heating power is small, and the additional heat generated can be completely diffused, so the temperature of the battery remains basically unchanged. The voltage characteristics of the battery can be detected on a long-term scale. The development of short circuits during this stage is slow, occupying most of the entire evolution process. Inspection during this stage is difficult and often can only be achieved by observing the voltage characteristics of the battery.

Middle stage: After a period of development, the resistance of the internal short circuit decreases, and the self-discharge current gradually increases, accompanied by an increase in heating power and an increase in voltage drop rate. At this point, the heat generated by the internal short circuit cannot be dissipated by the battery in a timely manner, resulting in a significant increase in battery temperature. At this stage, the heating and voltage changes in the battery are very obvious, and the heating will accelerate the aging of the battery, causing chemical reactions or decomposition between the electrode material and the electrolyte. The diaphragm will also be affected by temperature, resulting in performance degradation or even deformation. Therefore, the occurrence rate of short circuits during this stage is very fast, often in an exponential form over time. The internal short circuit at this stage can be evaluated through electrical and thermal characteristics, and it is relatively simple.

In the final stage, as the battery structure is damaged, the internal short-circuit resistance further decreases, resulting in a further increase in heating power. Due to the increase in heat generation, the temperature of the battery further rises. When it reaches a specific limit, the separator decomposes and collapses, causing extensive contact between the positive and negative electrodes of the battery. This leads to a sudden increase in

heating power, triggering reactions and decomposition between the electrode material and the electrolyte depth, and causing thermal runaway of the battery. At this stage, the voltage of the battery will experience a significant drop, even to 0 V, and energy will be released instantly, often leading to fire and explosion, with very obvious characteristics. The internal short circuit at this stage is no longer meaningful for detection.

2.3. Analysis of the ISC Process Principle

During the use of lithium batteries, electronic pathways are formed inside, leading to internal short circuits. During the manufacturing process, if metal impurities are accidentally mixed into the electrode material, or if burrs are formed on the edges of the current collector due to cutting, internal short circuits may occur [26]. During the service process, lithium batteries usually face complex and diverse environments. Charging at low temperatures or undergoing high-rate charging can lead to lithium precipitation and growth at the negative electrode of the battery. If it pierces the separator, it will form an internal short circuit [27]. As Figure 5 shows, the mechanism process of internal short circuit caused by overcharging in lithium batteries, Figure 5a shows overcharge, Figure 5b shows SEI film thickening, Figure 5c shows lithium plating with deformation, and Figure 5d shows ISC occurrence. Overcharging can cause structural damage or even collapse of the positive electrode active material, accelerating the aging rate of the battery while increasing the internal resistance, resulting in an increase in Joule heat inside the battery and causing an increase in internal temperature. As Figure 5d shows, when lithium plating penetrate the separator, as shown in the red box of the figure, it will trigger a reaction between the electrolyte and the electrode material. When the temperature continues to rise to the tolerance value of the separator, the separator will shrink or dissolve, leading to internal short circuit [28–30].

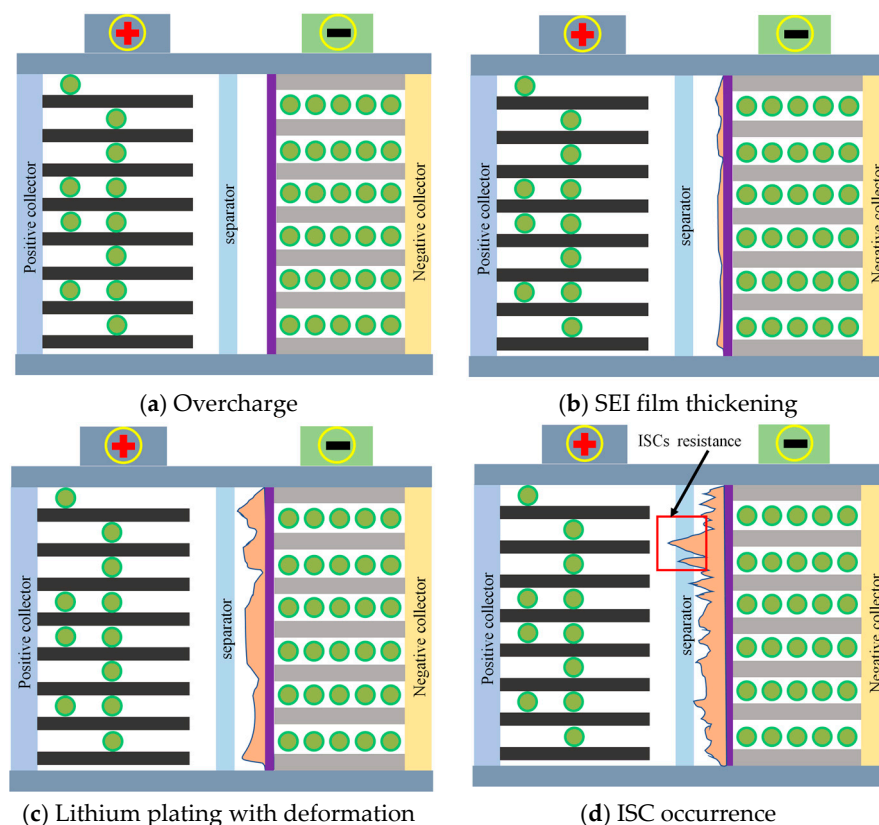


Figure 5. ISC process.

The external and internal short circuits of batteries have similarities in terms of thermal runaway paths. Both external and internal short circuits are triggered by low impedance paths, resulting in a sudden increase in current. At the same time, the abnormal characteristics of the voltage curve generated by external short circuits, such as sustained low voltage, are highly consistent with the mid-term evolution stage of internal short circuits [31,32], as shown in Figure 6a,b. Therefore, external short circuit experiments can be used to simulate the risk of internal short circuits and establish correlation models and safety assessment methods.

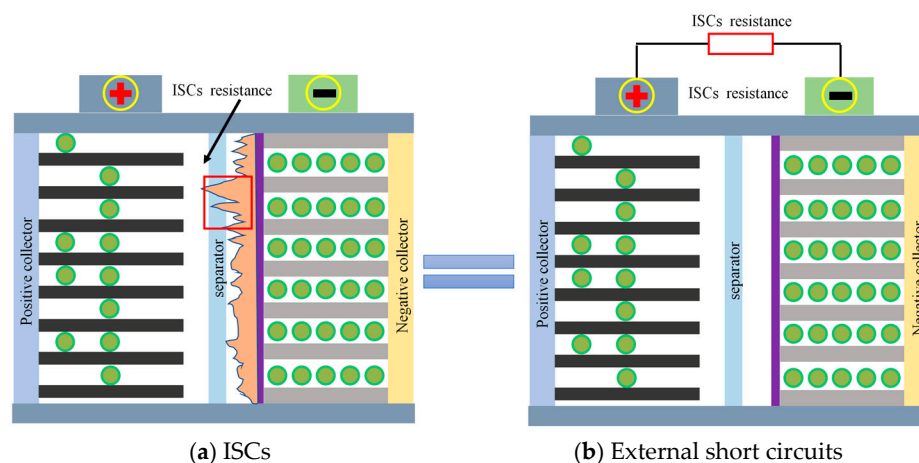


Figure 6. ISCs and external short circuits.

3. Coupling Model

3.1. Electrochemical Model

The gel-electrolyte lithium-ion batteries were composed of the positive electrode material and the negative electrode material. The positive electrode material is NCM811, the negative electrode material is graphite (Li_xC_6), and the electrolyte is composed of EC: DMC solvent with a volume ratio of 1:2, p (VdF HFP), and 2M LiPF_6 for simulation.

(1) Solid phase and liquid phase lithium ion diffusion process

Solid phase:

The mass balance of lithium ions in the active solid material particles of the battery is controlled by Fick's second law:

$$\frac{\partial c_{s,i}}{\partial t} = D_{s,i} \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial c_{s,i}}{\partial r} \right) \quad (1)$$

where $i = p$ is the negative electrode and $i = n$ is the positive electrode. $c_{s,i}$ is the concentration of Li^+ within the active electrode particle, $D_{s,i}$ is the diffusion coefficient of lithium-ions in the solid phase, $r \in (0, R_{s,i})$ is the radial coordinate of the active particle, and $R_{s,i}$ is the radius of the assumed spherical particle.

Boundary conditions:

$$\begin{cases} -D_{s,i} \frac{\partial c_{s,i}}{\partial r} \Big|_{r=0} = 0, (\text{particle center}) \\ -D_{s,i} \frac{\partial c_{s,i}}{\partial r} \Big|_{r=R_{s,i}} = \frac{j_n}{a_s F}, (\text{particle surface}) \end{cases} \quad (2)$$

where j_n is the local electrofluid density, a_s is the specific surface area of the active electrode particles, and F is the Faraday constant.

Liquid phase:

The material balance of the electrolyte in the liquid phase is as follows:

$$\varepsilon_i \frac{\partial c_i}{\partial t} = D_i^{eff} \frac{\partial^2 c_i}{\partial x^2} - \frac{i_i \cdot \nabla t_+}{F} + \frac{(1 - t_+)}{F} j_n, (i = p, s, n) \quad (3)$$

where $i = s$ is the separator and the pore wall flux equals zero at the separator.

Boundary conditions:

$$-D_p^{eff} \frac{\partial c_p}{\partial x} \Big|_{x=0} = 0, -D_n^{eff} \frac{\partial c_n}{\partial x} \Big|_{x=L} = 0 \quad (4)$$

where ε_i is the volume fractions of the negative electrode, electrolyte, and positive electrode. $D_i^{eff} = D_i(\varepsilon_i)^{Brugg}$ is the effective diffusion coefficient of the liquid phase. x is the transverse coordinate variable. t_+ is the Li^+ transference number, and $L_n + L_s + L_p = L$ is the sum of the thicknesses of the positive electrode, negative electrode, and separator layers.

The concentration and flux of the electrolyte are continuous, as shown below.

$$\left\{ \begin{array}{l} c_p \Big|_{x=L_p^-} = c_s \Big|_{x=L_p^+} \\ c_s \Big|_{x=(L_p+L_s)^-} = c_n \Big|_{x=(L_p+L_s)^+} \\ -D_p^{eff} \frac{\partial c_p}{\partial x} \Big|_{x=L_p^-} = -D_s^{eff} \frac{\partial c_s}{\partial x} \Big|_{x=L_p^+} \\ -D_s^{eff} \frac{\partial c_s}{\partial x} \Big|_{x=(L_p+L_s)^-} = -D_n^{eff} \frac{\partial c_n}{\partial x} \Big|_{x=(L_p+L_s)^+} \end{array} \right. \quad (5)$$

For gel-electrolyte lithium-ion batteries, $t_+ = 0.6$, and gel-electrolytes exhibit lower ion diffusion coefficients compared to liquid-electrolytes, which significantly impacts lithium-ion transmission rates and polarization behavior.

For gel-electrolyte lithium-ion batteries, $D_i = 5 \times 10^{-11}$ (m²/s). The enhanced ion mobility in gel-electrolytes can restrict anion movement, thereby mitigating salt depletion under high current conditions.

For gel-electrolyte lithium-ion batteries, $\varepsilon_e = 0.1$. Gel-electrolytes fill and occupy electrode pores, leading to a reduction in effective liquid space within the electrode structure.

(2) Solid phase and liquid phase electric potential distribution

Solid phase:

The electric potential distribution in the solid phase is governed by Ohm's law:

$$\sigma_i^{eff} \frac{\partial^2 \phi_{s,i}}{\partial x^2} \Big|_{x=0} = j_n, (i = p, n) \quad (6)$$

Boundary conditions:

$$\left\{ \begin{array}{l} -\sigma_p^{eff} \frac{\partial \phi_{s,p}}{\partial x} \Big|_{x=0} = I_{app}, (collector \text{ to } negative \text{ electrode}) \\ -\sigma_p^{eff} \frac{\partial \phi_{s,p}}{\partial x} \Big|_{x=L_p} = -\sigma_n^{eff} \frac{\partial \phi_{s,n}}{\partial x} \Big|_{x=L_p+L_s} = 0, (electrode \text{ to } separator) \end{array} \right. \quad (7)$$

where I_{app} is the current density of the battery.

For gel-electrolyte lithium-ion batteries, the potential of the solid phase at the positive electrode of the battery is set to zero.

$$\phi_{s,n} \Big|_{x=L_p+L_s+L_n} = 0 \quad (8)$$

liquid phase:

The expression for the charge balance in the battery based on Ohm's law is as follows:

$$\begin{cases} j_n = -\frac{\partial}{\partial x}(\kappa_i^{eff} \nabla \phi_{e,i}) + \frac{2RT\kappa_i^{eff}}{F}(1-t_+)(1 + \frac{\partial \ln f_{\pm}}{\partial \ln c_{e,i}}) \frac{\partial}{\partial x}(\frac{\partial \ln c_{e,i}}{\partial x}), (i = p, s, n) \\ \kappa_i^{eff} = \kappa \epsilon_i^{Brugg} \end{cases} \quad (9)$$

where κ_i^{eff} is the effective Li^+ conductivity, R is the gas constant, T is the temperature of the lithium-ion battery, $f_{\pm}$ is the molar activity coefficient, and κ is the Li^+ conductivity in the liquid phase.

For gel-electrolyte lithium-ion batteries, at the terminals of the battery, there is no charge flux in the liquid phase:

$$-\kappa_p^{eff} \frac{\partial \phi_{e,p}}{\partial x} \Big|_{x=0} = -\kappa_n^{eff} \frac{\partial \phi_{e,n}}{\partial x} \Big|_{x=L_p+L_s+L_n} = 0 \quad (10)$$

(3) Solid phase and liquid phase interface reaction

Behavior at these interfaces is determined by the Butler–Volmer equation:

$$j_n = a_s i_0 \left[\exp\left(\frac{\alpha_n F}{RT} \eta\right) - \exp\left(-\frac{\alpha_p F}{RT} \eta\right) \right] \quad (11)$$

where i_0 is the exchange current density, α_n is the transfer coefficient of the electrochemical reaction at the positive electrode, α_p is the transfer coefficient of the electrochemical reaction at the negative electrode, which is typically taken as 0.5, and η is the overpotential.

For gel-electrolyte lithium-ion batteries, i_0 depends on the lithium concentration in the electrolyte and the solid active substance, which is as follows:

$$i_0 = F(k_p)^{\alpha_n} (k_n)^{\alpha_p} (c_{s,\max} - c_s)^{\alpha_n} (c_s)^{\alpha_p} \left(\frac{c_e}{c_{e,ref}}\right)^{\alpha_n} \quad (12)$$

where $k_p = k_n = 0.04$ (m/s). The reaction rate constants for gel-electrolytes are typically one order of magnitude lower than those of liquid-electrolytes.

(4) Battery terminal voltage

Battery terminal voltage $V(t)$ can be expressed as:

$$V(t) = \phi_s|_{x=L} - \phi_s|_{x=0} \quad (13)$$

3.2. Thermal Model

The internal heat sources of gel-electrolyte lithium-ion batteries mainly comprise reversible heat, irreversible heat, and heat generated from ISCs. Battery heat energy conservation equation:

$$\rho_c c_{pc} \frac{\partial T}{\partial t} = \nabla \cdot k_c \nabla T + Q_{irr} + Q_{rev} + Q_{ohm} + Q_a \quad (14)$$

where ρ_c and c_{pc} are respectively the density of the battery and the specific heat capacity. T is the battery temperature. k_c is the thermal conductivity of the battery. Q_{irr} is irreversible heat. Q_{rev} is reversible heat generation. Q_{ohm} is ohm heat. Q_a is the secondary heat exchanger.

For gel-electrolyte lithium-ion batteries, $c_{pc} = 2$ (J/(kg·K)). Gel-electrolyte lithium-ion batteries have higher specific heat capacity than liquid-electrolyte batteries.

Due to the multilayer structure of gel-electrolyte lithium-ion battery, its thermal conductivity varies in different directions, where L_i is the thickness of each layer:

$$\begin{cases} k_{c,x} = \frac{|x|}{r} \frac{\sum L_i}{\sum L_i/k_{c,i}} + \frac{|y|}{r} \frac{\sum L_i k_{c,i}}{\sum L_i} \\ k_{c,y} = \frac{|y|}{r} \frac{\sum L_i}{\sum L_i/k_{c,i}} + \frac{|x|}{r} \frac{\sum L_i k_{c,i}}{\sum L_i} \\ k_{c,z} = \frac{\sum L_i k_{c,i}}{\sum L_i} \end{cases} \quad (15)$$

$$\begin{cases} Q_{irr} = J_n(\phi_s - \phi_e - U_{ref}(\theta)) \\ Q_{rev} = J_n * T * \frac{\partial U_{ref}(\theta)}{\partial T} \\ Q_{ohm} = \sigma^{eff} \nabla \phi_s \cdot \nabla \phi_s + (k^{eff} \nabla \phi_e \cdot \nabla \phi_e + \frac{2RTk^{eff}}{F} (1 + \frac{\partial \ln f_{\pm}}{\partial \ln c_e}) \nabla \ln c_e \cdot \nabla \phi_e) \end{cases} \quad (16)$$

The heat generation equation for gel-electrolyte lithium-ion battery side reactions is established based on the Arrhenius equation. the total heat generation from side reactions, Q_a can be expressed as follows:

$$Q_a = Q_{sei} + Q_{ne} + Q_{pe} + Q_e \quad (17)$$

where Q_{sei} is the SEI decomposition heat, Q_{ne} is the negative electrode and the electrolyte, Q_{pe} is positive electrode and the electrolyte reaction, and Q_e is decomposition of the electrolyte.

The metastable components of the SEI layer begin to exothermically decompose at approximately 90 °C. The reaction rate R_{sei} of this exothermic decomposition reaction can be expressed as follows:

$$R_{sei} = A_{sei} \exp\left(-\frac{E_{a,sei}}{RT}\right) c_{sei}^{m_{sei}} \quad (18)$$

where A_{sei} is the frequency factor for SEI decomposition; $E_{a,sei}$ is the activation energy of the reaction; c_{sei} is the dimensionless concentration of the metastable lithium-containing species within the SEI layer; and m_{sei} is the reaction order.

Q_{sei} is side reaction heat of SEI decomposition, and C_{sei} is the change rate, which can be expressed as follows:

$$\begin{cases} Q_{sei} = H_{sei} W_c R_{sei} \\ \frac{dc_{sei}}{dt} = -R_{sei} \end{cases} \quad (19)$$

where H_{sei} is specific heat release, and W_c is specific carbon content.

Negative reaction:

At temperatures exceeding 120 °C, the intercalated lithium within the negative electrode interacts with the gel-electrolyte. The rate R_{ne} of negative electrode reaction is formulated as follows:

$$R_{ne} = A_{ne} \exp\left(-\frac{t_{sei}}{t_{sei,ref}}\right) \exp\left(-\frac{E_{a,ne}}{RT}\right) c_{ne}^{m_{ne}} \quad (20)$$

where A_{ne} is the decomposition frequency factor, $E_{a,ne}$ is the activation energy of the reaction, t_{sei} is the thickness of SEI layer, c_{ne} is dimensionless concentration of lithium in the negative electrode, and m_{ne} is the reaction order.

For gel-electrolyte lithium-ion batteries, $A_{ne} = 2 \times 10^{10}$ (S⁻¹). Gel-electrolyte lithium-ion batteries reduce this factor by 1–2 orders of magnitude, blocking interface contact and mass transfer.

Positive reaction:

At more than 170 °C, the positive has an exothermic decomposition reaction due to accepting a higher temperature. And then the released oxygen directly reacts with the battery electrode to release a lot of heat. The positive reaction rate R_{pe} is as follows:

$$R_{pe} = A_{pe} \alpha^{m_{pe}} (1 - \alpha)^{m_{pe}} \exp\left(-\frac{E_{a,pe}}{RT}\right) \quad (21)$$

where A_{pe} is the decomposition frequency factor, $E_{a,pe}$ is the activation energy of the reaction, α is the conversion rate of positive active substance, and m_{pe} is the reaction order.

For gel-electrolyte lithium-ion batteries, $A_{pe} = 3.5 \times 10^{13} \text{ (S}^{-1}\text{)}$. Gel-electrolyte lithium-ion batteries reduce this factor by 1–3 orders of magnitude, crucial for restraining thermal runaway.

Gel-electrolyte decomposition:

The decomposition of the remaining electrolyte usually begins above 250 °C, and the reaction rate R_e is as follows:

$$R_e = A_e \exp\left(-\frac{E_{a,e}}{RT}\right) c_e^{m_e} \quad (22)$$

where A_e is the decomposition frequency factor, $E_{a,e}$ is the activation energy of the reaction, c_e is the dimensionless concentration of electrolyte, and m_e is the reaction order.

For gel-electrolyte lithium-ion batteries, $A_e = 5.14 \times 10^{25} \text{ (S}^{-1}\text{)}$. Gel-electrolyte lithium-ion batteries have a greatly reduced decomposition frequency factor due to the polymer network binding reactants and slowing PEO and other polymer thermal decomposition.

For gel-electrolyte lithium-ion batteries, $E_{a,e} = 300 \text{ (kJ/mol)}$. Gel-electrolyte lithium-ion batteries require additional energy to break polymer covalent bonds, increasing apparent activation energy.

Convection heat transfer and radiation heat transfer on the battery surface are considered, and the following relationships are given.

$$\begin{cases} q_{conv} = h(T_s - T_{amb}) \\ q_{rad} = \varepsilon \sigma (T_s^4 - T_{amb}^4) \end{cases} \quad (23)$$

where q_{conv} and q_{rad} are the convective heat flux at boundary and the radiative heat flux and T_s and T_{amb} are battery surface temperature and the ambient temperature. h is the convection heat transfer coefficient, ε is the radiation coefficient of the lithium battery surface, and σ is the Stefan-Boltzmann constant.

3.3. ISC Model

At different ISC situations, short-circuit current path or thermal path may vary, and so it is difficult to clarify the mechanism. However, different ISCs share common characteristics: the electron current flows from the positive collector to the negative collector, and the heat generated during the short circuit accumulates in the vicinity of the short circuit region before dissipating outward. Based on these observations, the short circuit can be simulated by introducing a short circuit region, as shown in Figure 7.

The short-circuit region of the battery is conductive. The current passing through the short-circuit region can vary depending on the resistance. If the resistance of the short-circuit area is known, the short-circuit current and Joule heating generated are as follows:

$$I_{short} = \iiint_{n.or.p} J_n dV \quad (24)$$

where I_{short} is the battery short-circuit current, n and p are, respectively, the positive electrode and the negative electrode, and v is the battery volume.

$$Q_{short} = I_{short}^2 R_{short} \quad (25)$$

where Q_{short} is joule heating generated by a short circuit in the battery; R_{short} is the short circuit resistance in the battery.

As a result, the electron current between the active materials and the short-circuit region can be considered negligible. Under these conditions, the short circuit resistance is as follow.

$$R_{short} = \frac{L_{short}}{\sigma_{short} \pi r_{short}^2} \quad (26)$$

where L_{short} is length of the short circuit region, σ_{short} is the conductivity of short-circuit region, and r_{short} is the radius of the region.

For gel-electrolyte lithium-ion batteries, the magnitude of the short-circuit resistance serves as an indicator of whether the battery has undergone a significant ISC. When ISC failures occur, key parameters such as voltage, current, temperature, SOC, and capacity will also exhibit corresponding changes.

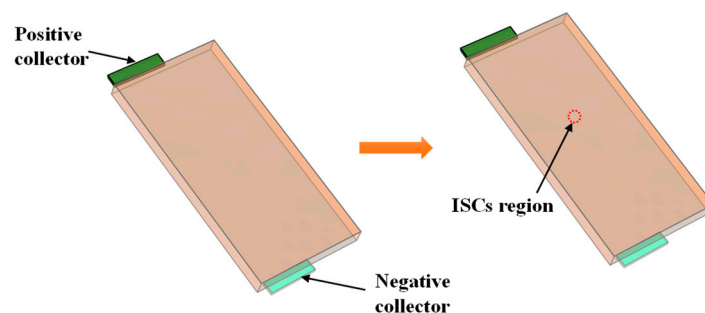


Figure 7. Battery ISC structure.

3.4. Electrochemical–Thermal–ISC Coupling Model

A coupling relationship among the electrochemical, thermal, and ISCs characteristics of lithium-ion batteries has been established, as shown in Figure 8.

When ISC failures occur, a significant amount of heat is released internally. The internal resistance of the battery during the short circuit decreases. Consequently, a coupling relationship emerges between the electrochemical and thermal models.

$$U_p^{ref} = Eeq_int1(soc) + dEeqdT_int1(soc) * (T - 298[K]) \quad (27)$$

$$U_n^{ref} = Eeq_int2(soc) + dEeqdT_int2(soc) * (T - 298[K]) \quad (28)$$

where Eeq_int1 is the positive initial equilibrium potential, SOC is the battery charged state, $dEeqdT_int1$ is the positive equilibrium potential temperature derivative, Eeq_int2 is the negative initial equilibrium potential, and $dEeqdT_int1$ is the negative equilibrium potential temperature derivative.

$$\begin{cases} D_n = 1.4523 \times 10^{-13} \times e^{\left(\frac{68025.7}{8.314 \times \left(\frac{1}{318[K]} - \frac{1}{T}\right)}\right)} \\ D_p = 5 \times 10^{-13} \times e^{\left(1200 \times \left(\frac{1}{298[K]} - \frac{1}{T}\right)\right)} \\ D_{ele} = DL_int3(c) \times e^{\left(\frac{16500}{8.314 \times \left(\frac{1}{298[K]} - \frac{1}{T}\right)}\right)} \end{cases} \quad (29)$$

where D_n is negative electrode, D_p is positive electrode, D_{ele} is electrolyte diffusion coefficient, DL_{int3} is the electrolyte interpolation function, and the parameters are derived from the finite element simulation software.

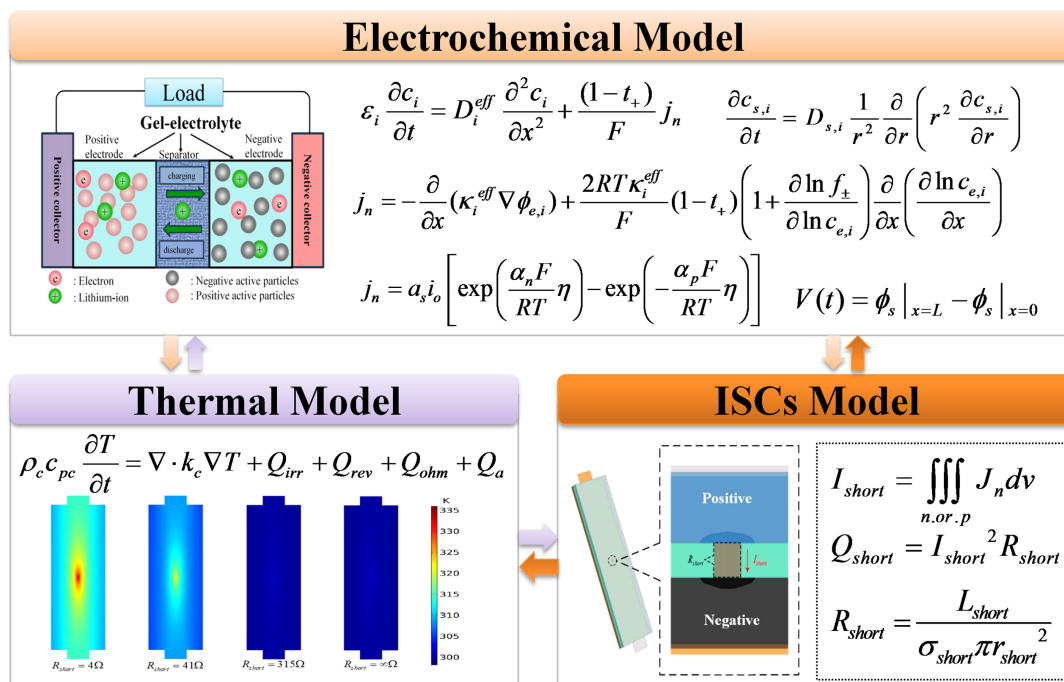


Figure 8. Electrochemical–thermal–ISC coupling.

4. Model Validation

Experimentally measuring the ISC characteristics of gel-electrolyte lithium-ion batteries validates ISC model accuracy. Battery is gel-electrolyte lithium-ion batteries, with positive materials— $\text{Li}[\text{Ni}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}]\text{O}_2$ —and negative materials— Li_xC_6 . The schematic diagram of the ISC experiment is shown in Figure 9.

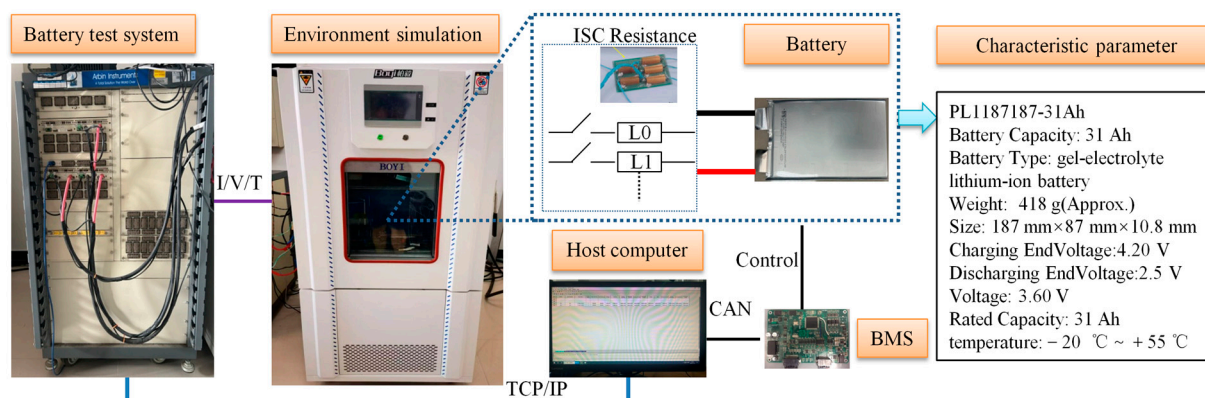


Figure 9. Experimental.

In order to verify the accuracy of the model and ensure the time and safety of the test, the test is carried out under the short-circuit resistance of 315 Ω . Battery voltage at four discharge multipliers is measured: 0.18 C, 0.33 C, 0.5 C and 1 C. Battery discharge voltage curve with different rates based on ISC model is simulated. The curves of simulation and test battery voltage as well as the comparative error curves are shown in Figure 10. The error is basically within 40 mV.

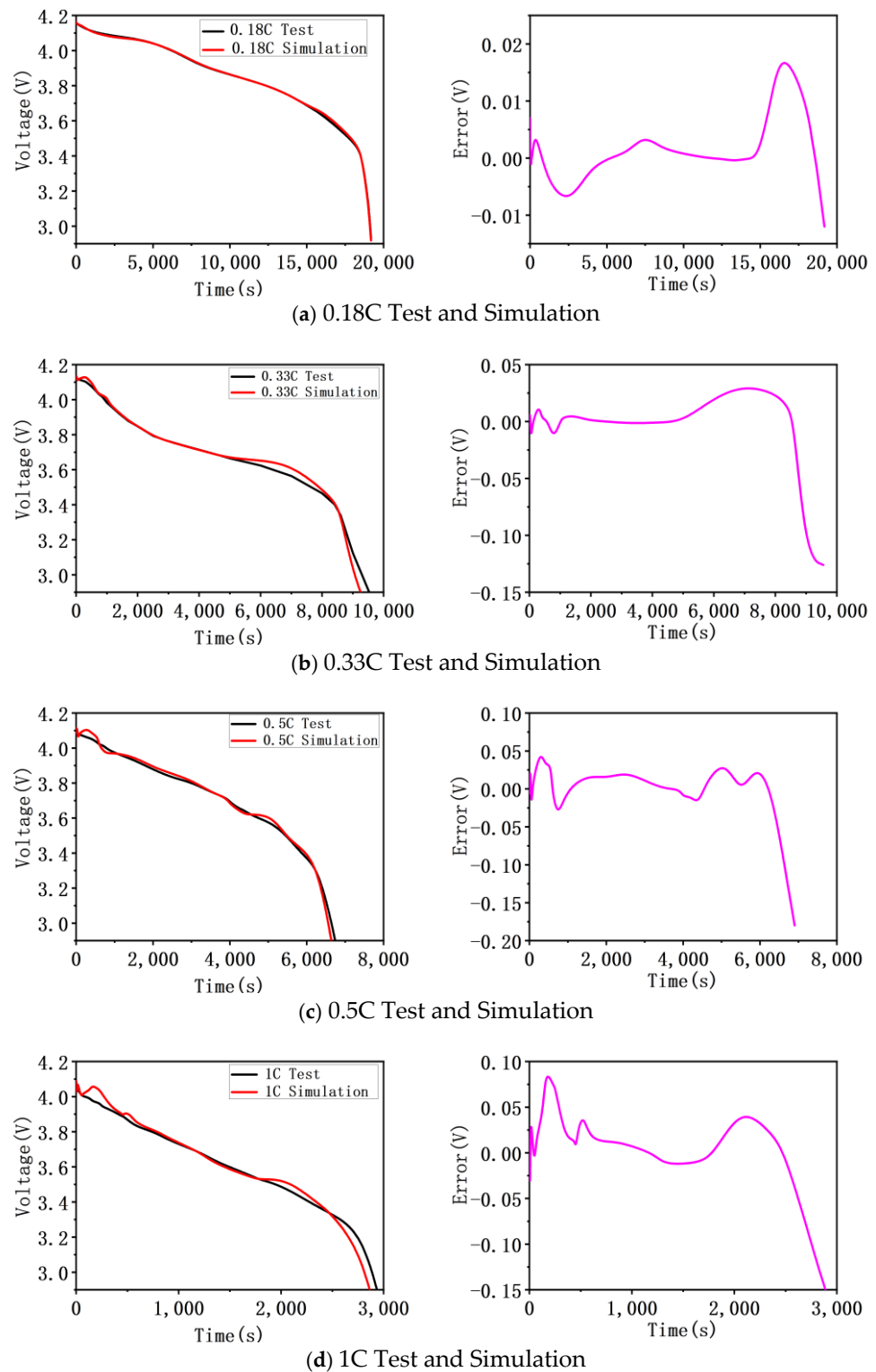


Figure 10. Simulation and test battery voltage.

Measurement of battery voltage at 1 C. Simulation of battery discharge temperature with 1 C rate based on ISC model. The curves of simulation and test battery voltage as well as the comparative error curves are shown in Figure 11. The error is basically within 0.5 °C.

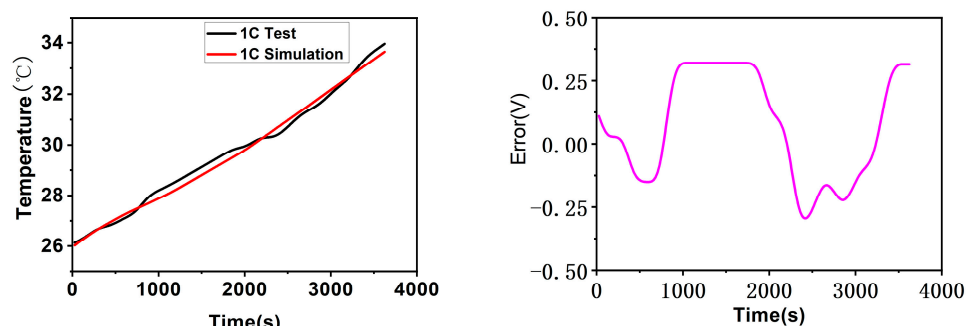


Figure 11. Simulation and test battery temperature.

Calculate the MAE of simulation voltage and test voltage, and simulation temperature and test temperature separately. The maximum voltage MAE is 25 mv, and the temperature MAE is 0.22 °C, as shown in Table 1.

Table 1. Voltage and temperature MAE.

Type	Charge/0.18 C	Charge/0.33 C	Charge/0.5 C	Charge/1 C	Temperature/1 C
MAE	0.0034 V	0.018 V	0.021 V	0.025 V	0.22 °C

5. Datasets Acquisition

The classification boundaries for ISCs in lithium-ion batteries are critical for the detection and diagnosis of battery faults [15]. Select R_{short} equal to 315 Ω , 41 Ω and 4 Ω as the critical points for the severity of internal short circuits. At $R_{\text{short}} = 315 \Omega$, the battery loses 15% of its power during self-discharge for 28 days. At $R_{\text{short}} = 41 \Omega$, the battery loses 100% of its power due to self-discharge over a period of 28 days. At $R_{\text{short}} = 4 \Omega$, the battery loses 100% of its power due to self-discharge over a period of 7 days. The critical points for short circuit classification are shown in Figure 12.

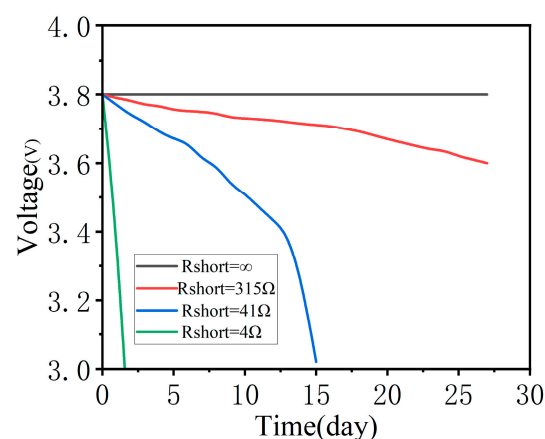


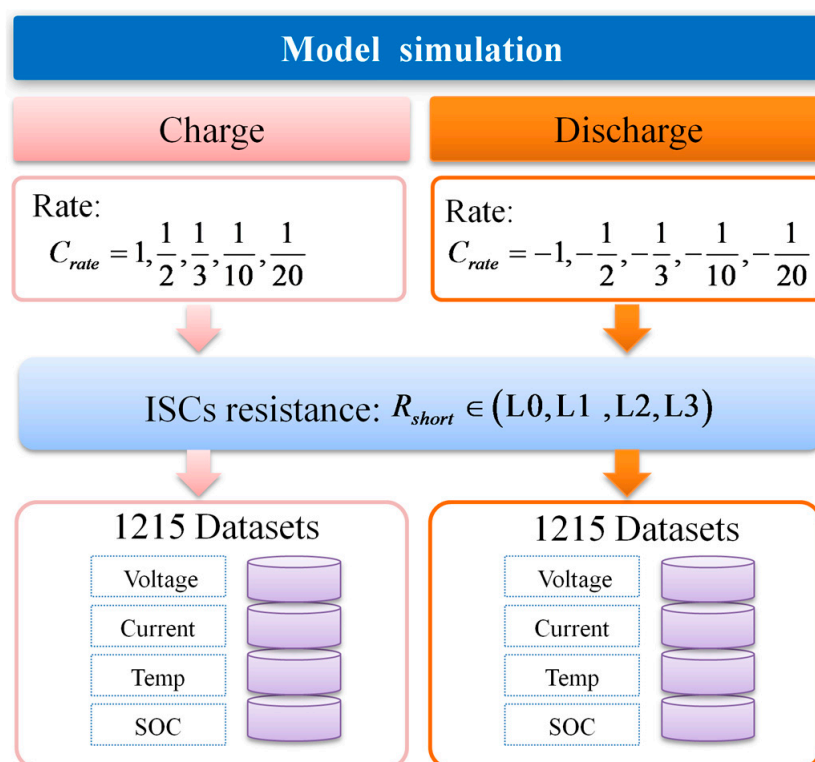
Figure 12. Battery characteristics in case of internal short circuit.

In this paper, four distinct levels are defined: L0, L1, L2, and L3, representing normal condition, Class I, Class II, and Class III severity, respectively. The higher the level, the more severe the ISCs. The level classifications are shown in Table 2.

Table 2. ISC level.

ISC R_{short} Range (Ω)	Level
Up to 315	L0
41~315	L1
4~41	L2
0~4	L3

Based on ISC model, a parameterized scanning simulation acquires a total of 1215 datasets of charge/discharge, which include voltage (V), current (A), temperature (T), and SOC. ISC dataset acquisition structure diagram is shown in Figure 13.

**Figure 13.** ISC datasets acquisition structure diagram.

6. Methods for ISC Prediction and Results

6.1. BP-CNN-LSTM ISC Prediction Model

BP ISC prediction model includes input layer, output layer, and hidden layer, and there is weight control between each node between each layer. CNN ISC prediction model includes input layer, convolutional layer, fully connected layer, and an output layer. LSTM ISC prediction model stores long-term information through a cell state and uses input gates, output gates, and forget gates to control information updates, forgetting, and output. The internal structure of prediction model is shown in Figure 14.

In this paper, the charge and discharge dataset is divided into two parts, including training dataset and validation dataset. The parameters of model are shown in Table 3.

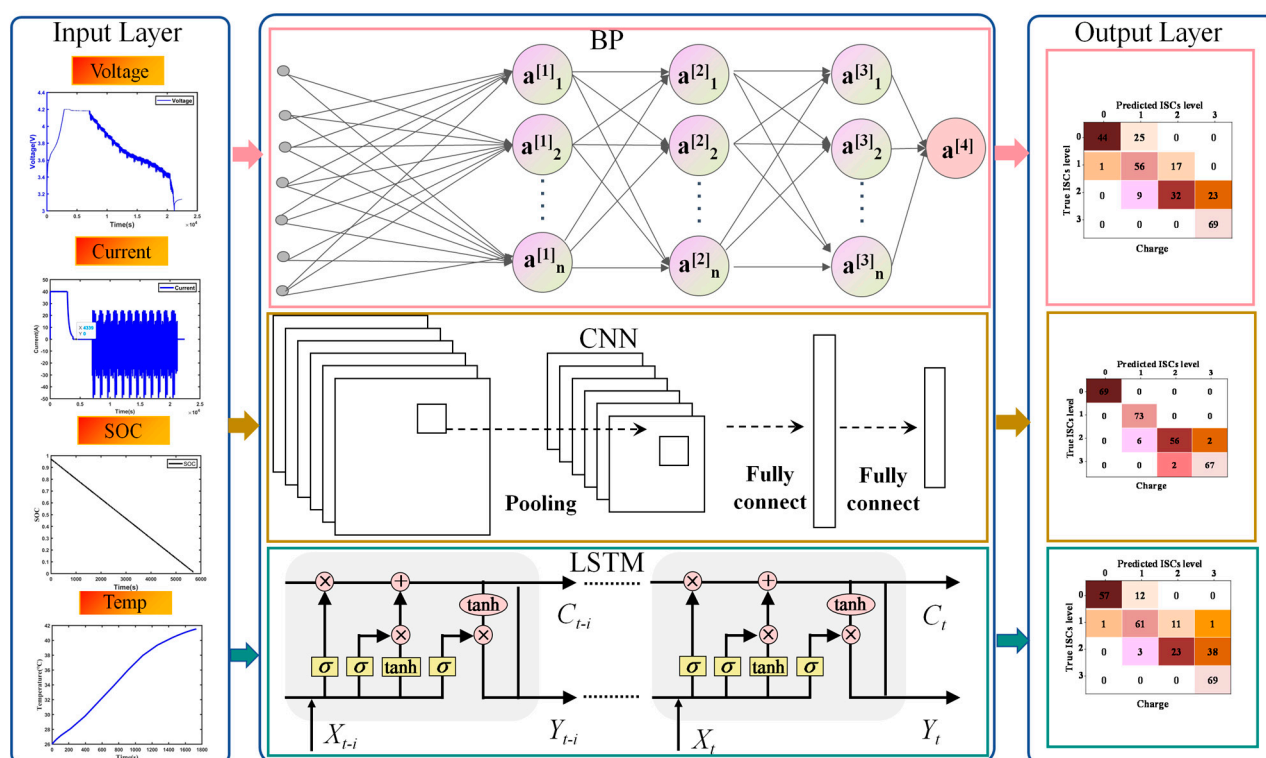


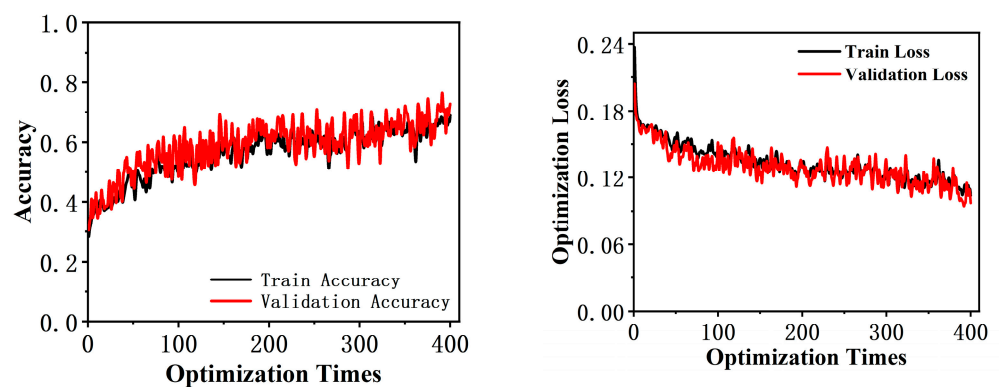
Figure 14. The internal structure of prediction model.

Table 3. Prediction model parameters.

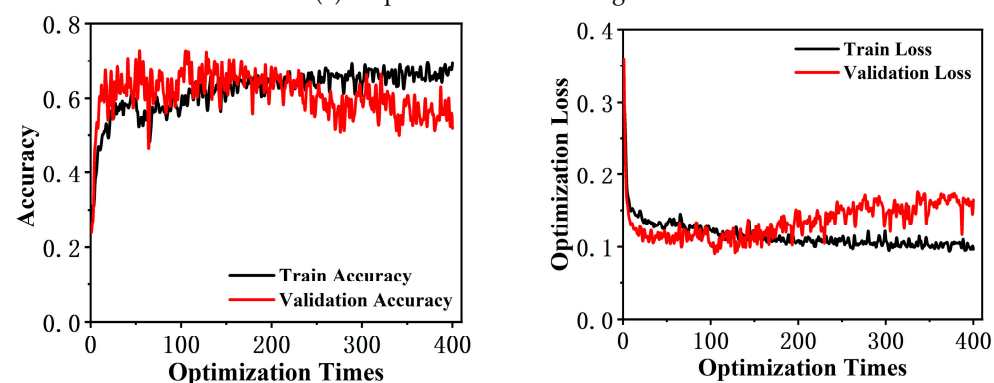
BP	Value	CNN	Value	LSTM	Value
training dataset	939	training dataset	939	training dataset	939
validation dataset	276	validation dataset	276	validation dataset	276
layers	2	layers	4	layers	5
input neurons	1836	input neurons	1836	input neurons	2115
output neurons	4	output neurons	4	output neurons	4
Loss function	SCC	Loss function	MSE	Loss function	SCC
Optimizer	Adam	Optimizer	Adam	Optimizer	Adam
Learning rate	1×10^{-3}	Learning Rate	1×10^{-3}	Learning Rate	1×10^{-4}
Batch size	32	Batch size	32	Batch size	20
Training cycle	400	Training cycle	400	Training cycle	400

6.2. Prediction Results

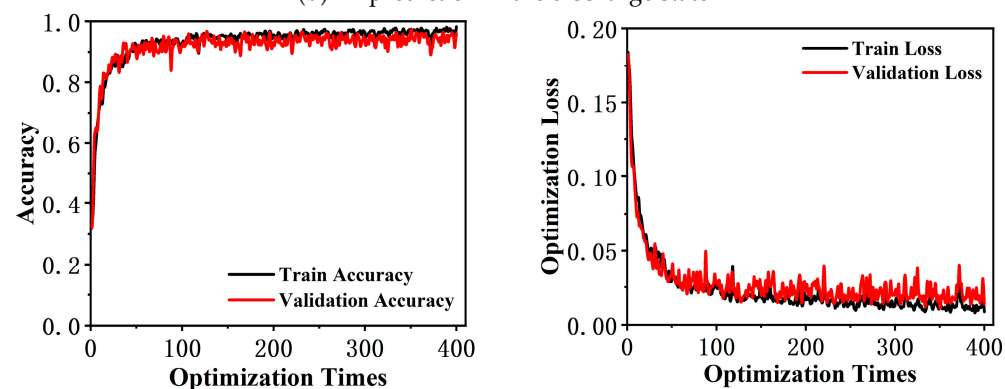
The prediction algorithm is divided into charging and discharging states. Input data include voltage, current, temperature, and SOC. Output data are predicted ISC levels. The accuracy and optimization loss are shown in Figure 15. The predicted ISC level classifications are shown in Figure 16.



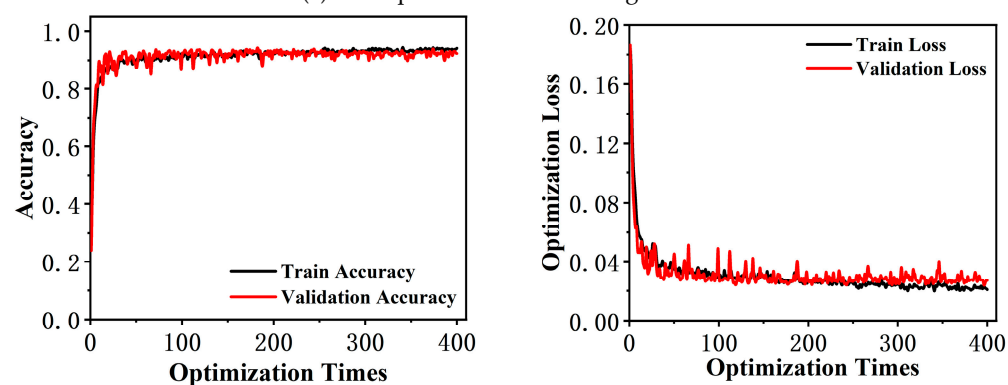
(a) BP prediction in the charge state



(b) BP prediction in the discharge state



(c) CNN prediction in the charge state



(d) CNN prediction in the discharged state

Figure 15. Cont.

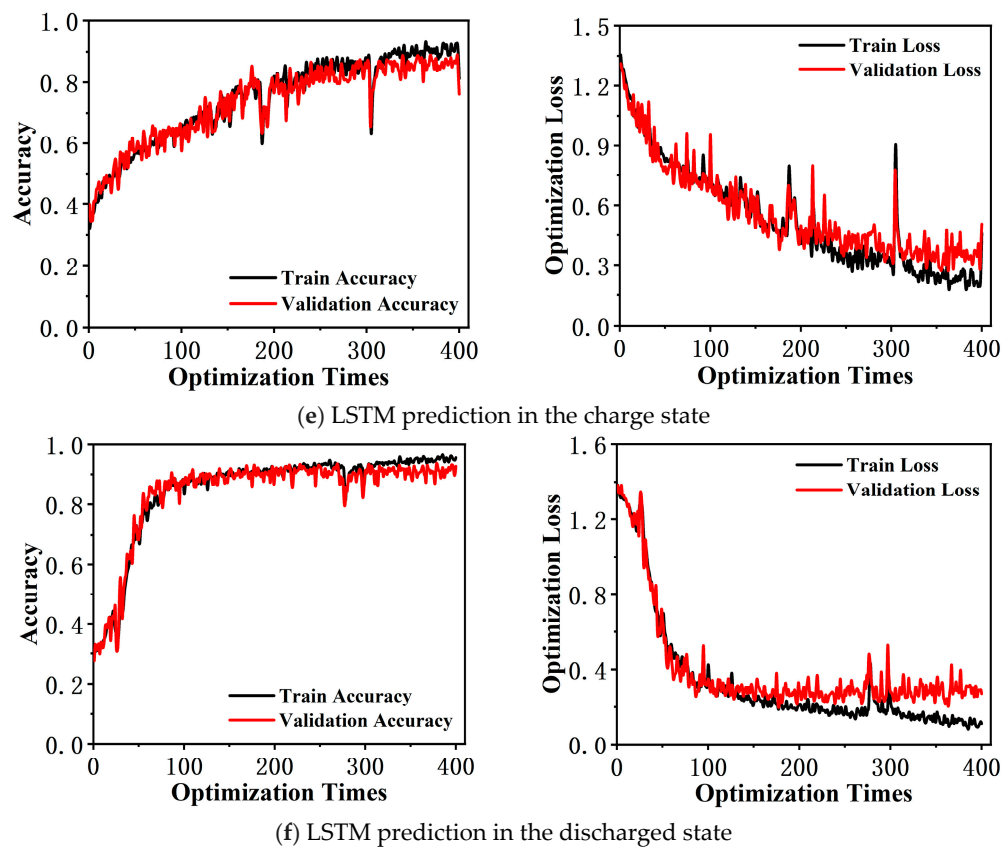


Figure 15. The accuracy and optimization loss.

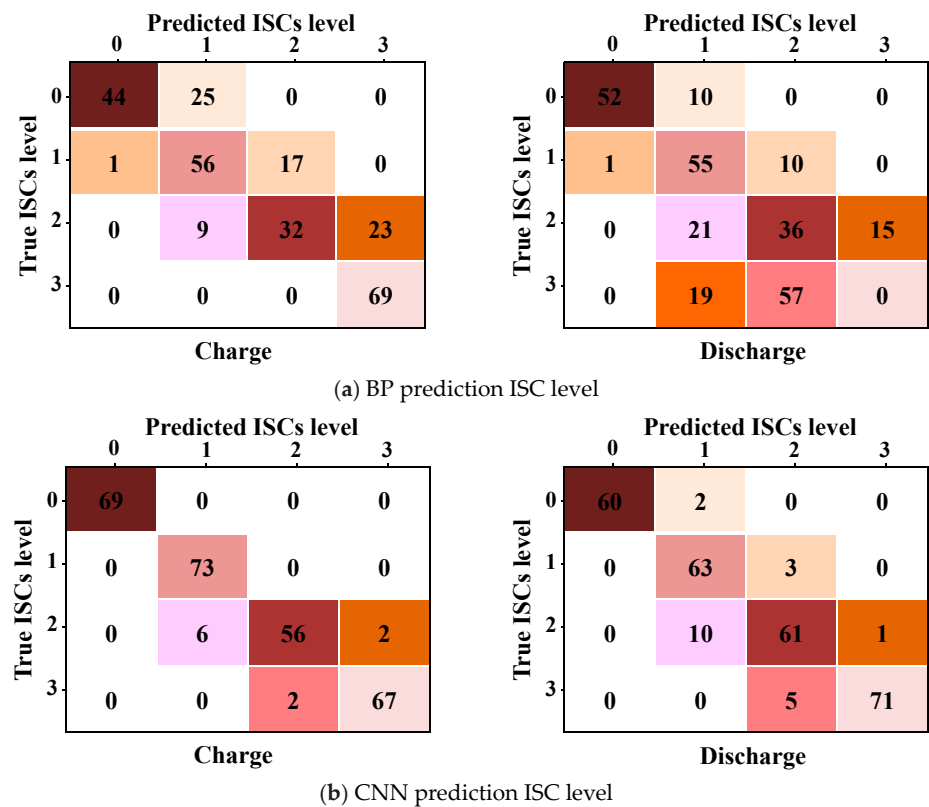


Figure 16. Cont.

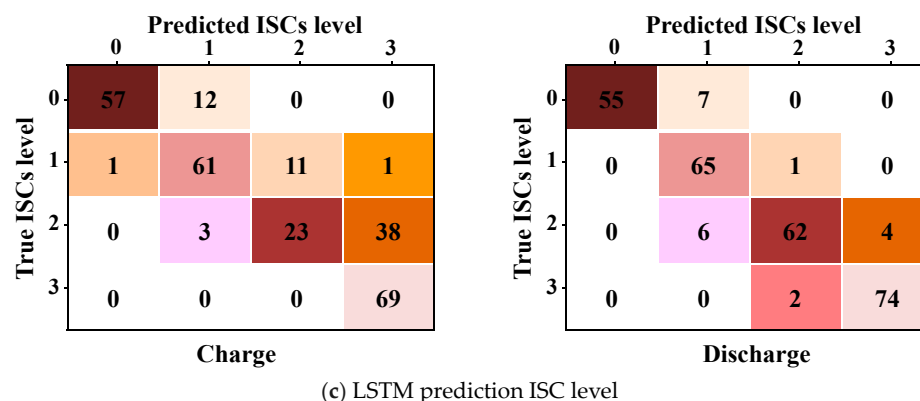


Figure 16. Predicted ISC level.

6.3. Discussion

In order to accurately and quantitatively compare ISC prediction algorithms, four evaluation parameters, including model accuracy, precision, recall, and F-value, were used for comparison. Firstly, the test set is defined as $\Phi = \{(x^{(1)}, y^{(1)}), \dots, (x^{(276)}, y^{(276)})\}$, with the internal short-circuit level label $y^{(n)} \in \{L3, L2, L1, L0\}$ of the battery. The test set is input into the trained model $f(x, y)$ for testing and classification, and the diagnosed level result is $\{\hat{y}^{(1)}, \dots, \hat{y}^{(276)}\}$.

Accuracy refers to the average of the overall classification and prediction performance of the test set, calculated as follows:

$$\mathcal{A} = \frac{1}{276} \sum_{n=1}^{n=1} I(y^{(n)} = \hat{y}^{(n)}) \quad (30)$$

$I(\cdot)$ is the indicator function.

Precision and recall are used to evaluate the prediction capability of each category. For example, if the level is L1, the model classification results may have four types:

(1) True example: The true level of a test sample is L1, and the prediction result of the model is also L1. This test sample size is referred to as:

$$TP_{L1} = \sum_{n=1}^{n=1} I(y^{(n)} = \hat{y}^{(n)} = L1) \quad (31)$$

(2) False negative example: If the true level of a test sample is L1, but the model diagnoses a result that is not L1, then the sample size is recorded as:

$$FN_{L1} = \sum_{n=1}^{n=1} I(y^{(n)} = L1 \wedge \hat{y}^{(n)} \neq L1) \quad (32)$$

(3) False positive example: If the true level of a test sample is not L1, and the model diagnoses it as L1, then the sample size is recorded as:

$$FP_{L1} = \sum_{n=1}^{n=1} I(y^{(n)} \neq L1 \wedge \hat{y}^{(n)} = L1) \quad (33)$$

(4) True negative example: The true level of a test sample is not L1, and the prediction result of the model is also not L1. Such a test sample size is referred to as:

$$TN_{L1} = \sum_{n=1}^{n=1} I(y^{(n)} \neq L1 \wedge \hat{y}^{(n)} \neq L1) \quad (34)$$

The precision is expressed as:

$$P_{L1} = \frac{TP_{L1}}{TP_{L1} + FP_{L1}} \quad (35)$$

The recall is expressed as:

$$\mathfrak{R}_{L1} = \frac{TP_{L1}}{TP_{L1} + FN_{L1}} \quad (36)$$

The accuracy and recall of the prediction model for the severity of internal short circuits in batteries are calculated on an average basis for the overall category, expressed as:

$$P_{macro} = \frac{1}{4} \sum_{L_4}^{L=L_1} P_L \quad (37)$$

$$\mathfrak{R}_{macro} = \frac{1}{4} \sum_{L_4}^{L=L_1} R_c \quad (38)$$

In order to harmonize the average of the overall accuracy and recall of the model, the F-value evaluation parameter is used, expressed as:

$$F = \frac{(1 + \beta^2) P_{macro} \mathfrak{R}_{macro}}{P_{macro} + \mathfrak{R}_{macro}} \quad (39)$$

The results of the ISC prediction algorithm comparisons are shown in Table 4.

Table 4. Compared ISC prediction algorithms.

	BP		CNN		LSTM	
	CH	DIS	CH	DIS	CH	DIS
Accuracy (A)	72.8%	72.5%	96%	92.4%	76.1%	92.8%
Precision (P_{macro})	74.7%	84.8%	98.8%	97.7%	76.3%	95.3%
Recall ($\mathfrak{R}_{macro}$)	94.1%	84.1%	96.9%	94.8%	96.3%	97.2%
F-value (F)	82.9%	83.5%	97.8%	96.1%	83.8%	96.2%

It can be seen that all three prediction algorithms have good performance, and the CNN has more outstanding prediction ability, which is superior to the others prediction algorithms in various evaluation indicators, with better prediction performance in charge and discharge states.

7. Conclusions

By analyzing the mechanistic characteristics of gel-electrolyte lithium-ion batteries, an electrochemical–thermal–ISC coupling model was developed. Through performance characterization of ISC, several key parameters that best represent battery ISC behavior were selected, including voltage, current, temperature, SOC, capacity loss, and internal resistance. Subsequently, the datasets were preprocessed to meet training requirements. Then, BP, CNN, and LSTM prediction models were developed and compared for accuracy. The results demonstrate that CNN achieves higher accuracy than both BP and LSTM, and this accuracy has been validated through experimental testing. ISCs in batteries exhibit

potential, complexity, and danger, with a long-term development process. The main innovative contributions of this study are as follows.

- (1) The transport mechanism of lithium ions in gelled electrolytes was analyzed. Lithium ions combine with the polymer matrix, and under external force, the polymer chains move. The mutual movement between chains facilitates the directional transport of lithium ions, enabling the normal operation of lithium batteries.
- (2) To address the difficulty of developing and implementing a large number of internal short circuit experiments due to the poor repeatability and controllability of traditional battery internal short circuit experiments, a three-dimensional finite element electrochemical–thermal–internal short circuit coupling model for lithium-ion batteries was established. This model replaces real internal short circuit experiments through simulation testing.
- (3) ISC prediction model for lithium-ion batteries has been established. The classification level of the severity of internal short circuits in batteries has been defined. ISC prediction model can provide unique insights that traditional simulations find difficult to capture, especially when dealing with complex, dynamic, or high-dimensional problems. It can detect a simulated fault and this modeling approach could be applied to empirical data.

In future work, further consideration needs to be given to battery aging and the impact of actual complex working conditions. At the same time, in reality, batteries may experience multiple faults occurring simultaneously, and these fault characteristics are interrelated. High-precision estimation of ISCs can be achieved through the fusion of intelligent algorithms and hybrid models.

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