Lifetime Prediction of Direct Methanol Fuel Cells Based on Hybrid Predictive Modeling

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Abstract: Direct methanol fuel cells (DMFCs) face durability challenges that limit their widespread adoption. Traditional data-driven degradation models excel in short-term predictions but lack capability for long-term forecasting. To mitigate disturbances caused by the voltage rebound phenomenon in fuel cells, this paper proposes a hybrid prediction model that combines mechanistic modeling and machine learning. The core idea is to use mechanistic modeling to extract degradation-relevant internal impedance features from electrochemical impedance spectroscopy (EIS), and then leverage machine learning to predict the evolution of these features, thereby avoiding direct fitting of noisy voltage signals. Specifically, the model first fits EIS using a full-circuit equivalent circuit model (ECM) to extract key internal degradation parameters. It then combines these parameters with a convolutional neural network (CNN) to predict impedance parameter trends. Finally, by establishing a linear relationship between the predicted impedance and the output voltage, the remaining useful life (RUL) of the methanol fuel cell is reliably estimated. Comparison results between the hybrid prediction model, Gated Recurrent Unit (GRU) network, and iTransformer network show predicted failure threshold time errors of 1 hour, 11 hours, and 5 hours, respectively. These results demonstrate that the proposed hybrid model achieves the smallest time prediction error (1 hour), indicating superior performance for long-term prediction tasks. The hybrid model improves data tracking ability and prediction accuracy, and reduces dependence on large amounts of degradation data. This model provides a feasible solution for DMFC lifetime prediction.

Keywords: Direct Methanol Fuel Cell; Life Prediction; Machine Learning; Voltage Rebound Phenomenon; Electrochemical Impedance Spectroscopy

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1 Introduction

Proton exchange membrane fuel cells operate at low temperatures with minimal heat loss, making them suitable for automotive and portable applications [1]. However, high-pressure hydrogen fuel presents storage and safety concerns [2], accelerating development of liquid-fueled direct methanol fuel cells (DMFCs). As promising next-generation portable power sources, DMFCs offer high energy density, low cost, convenient fuel storage, and enhanced safety [3].

Fuel cell durability remains substantially below ideal levels [4], with widespread adoption constrained by limited durability and high maintenance costs [5]. Electrode corrosion, catalyst agglomeration, and membrane degradation constitute primary degradation mechanisms. Accurate aging modeling and remaining useful life (RUL) prediction enable proactive maintenance scheduling, extending system lifespan and enhancing operational reliability [6-8].

Data-driven approaches enable RUL prediction using external condition monitoring data without requiring comprehension of complex internal degradation mechanisms. Kamran et al. [9] proposed a sum-weighted wavelet-ex-

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treme learning machine model validated on Proton Exchange Membrane Fuel Cell (PEMFC) data from the PHM Challenge 2014. Ma et al. [10] employed long short-term memory (LSTM) networks for fuel cell performance prediction, achieving mean absolute percentage error of 0.0025. Vichard et al. [11] developed an echo state network degradation model with prediction horizons exceeding 2000 hours under dynamic loads. Mezzi et al. [12] incorporated Markov chains for load profile estimation, eliminating the need for a priori knowledge of future operating conditions.

Hybrid approaches combining model-based and data-driven methods address limitations of pure strategies. Research integrating particle filtering with LSTM [13] and extended Kalman filter with LSTM [14] has demonstrated enhanced prediction accuracy. Wang et al. [15] proposed a fusion strategy using electrochemical mechanism models to extract degradation metrics, achieving prediction horizons exceeding 60% of total useful life with 11.4% relative error.

This paper presents a hybrid prediction framework combining impedance-voltage mathematical modeling with convolutional neural network (CNN)-based impedance analysis. The approach addresses voltage rebound phenomena in DMFCs by incorporating internal electrochemical state information, enabling accurate degradation trend prediction and RUL estimation.

2 Data Sources

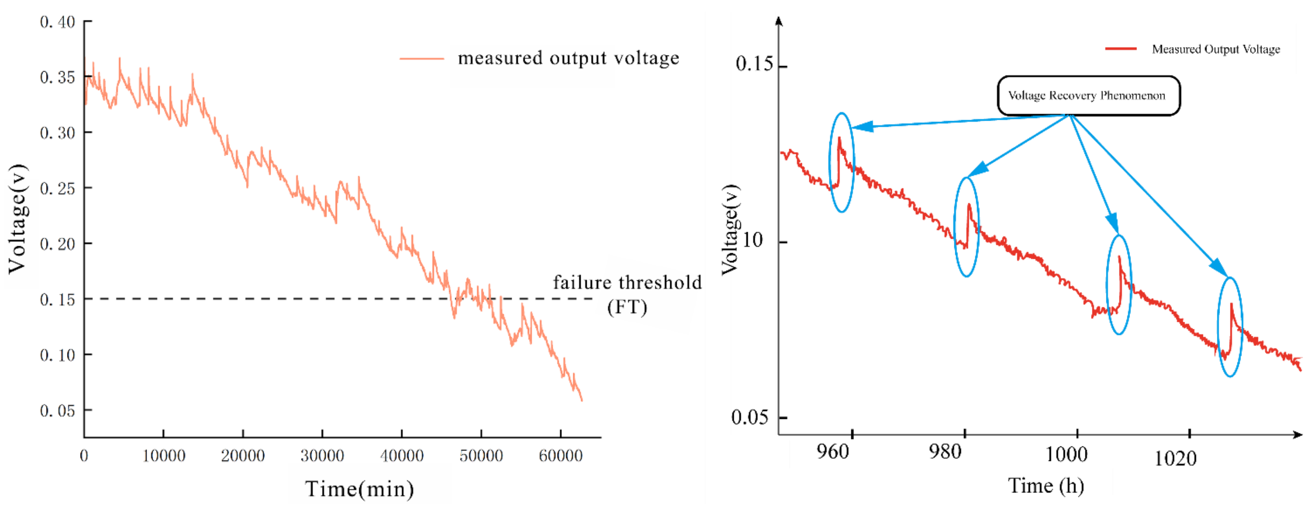
2.1 Experimental Platforms

The DMFC main body consists of cathode and anode end plates, current collector plates, and a membrane electrode assembly (MEA). The methanol cavity height is 15 mm, accommodating up to 4.86 ml of methanol solution. As a self-breathing DMFC, two through-holes were machined on the cavity sidewall to enable solution and gas exchange. A specialized miniature hot press maintained constant temperature and pressure during operation.

An ITECH IT8500 electronic load was used for extended discharge degradation testing, with current and voltage data acquired to construct polarization and power curves. These curves visually represent DMFC performance degradation. Due to inevitable factors such as manual preparation variations, membrane electrode quality differences, and ambient temperature and humidity fluctuations, DMFC performance exhibits non-negligible variability despite standardized preparation procedures. Discharge performance testing using an electronic load was conducted following activation to evaluate cell performance and select better-performing cells for subsequent long-term degradation testing.

2.2 Experimental Data Analysis

In DMFC degradation experiments, parameters such as temperature, pressure, and methanol concentration exhibit irregular variation patterns and significant acquisition uncertainty, making them difficult to employ directly as health indicators for cell performance degradation. Conversely, cell voltage data demonstrates clear trends during fuel cell degradation, intuitively reflecting performance. Consequently, voltage serves as a rapid assessment indicator for overall cell performance [16].



(a) Trend of actual output voltage of cell (b) Voltage recovery phenomenon

Figure 1 Cell voltage trends and recovery phenomenon

The DMFC degradation data presented in this paper were obtained from long-term constant-current discharge tests conducted on a single DMFC. The experiment lasted over 1000 hours, capturing changes in health indicators and experimental parameters. As evident from Figure 1(a), the voltage signal exhibits a significant decreasing trend, further confirming that cell output voltage can effectively characterize DMFC performance degradation. However, other experimental parameters show no obvious regular trend with time and thus cannot effectively characterize the fuel cell performance degradation process.

Based on voltage variations throughout the cell life cycle (Figure 1(b)), electrochemical impedance spectroscopy (EIS) measurements consistently induce a brief voltage increase followed by gradual decline—a phenomenon termed voltage rebound [17]. These rebound phenomena represent transient states that gradually diminish and disappear as operating conditions stabilize. Due to their stochastic nature in both magnitude and duration during DMFC degradation, voltage rebounds introduce difficulties for accurate lifetime prediction.

Fuel cell performance degradation primarily originates from aging of core components, processes typically difficult to monitor and observe directly. EIS, as a widely used non-invasive technique, assesses internal degradation processes by superimposing AC excitation at different frequencies on direct current [18]. Through EIS analysis and development of corresponding equivalent circuit models (ECM), impedance, capacitance, and other circuit component parameters can be fitted [19], facilitating extraction of degradation features that characterize the internal state.

2.3 Data Preprocessing

Due to the complexity and uncertainty of the experimental environment, collected operating parameter data may contain significant noise and spikes. The Savitzky-Golay (SG) filter is commonly applied to time series data for noise reduction while retaining main signal characteristics [20].

The SG filtering algorithm is a polynomial smoothing method based on the least squares principle. Given 2M+1 samples (centered at n=0), the SG filter performs polynomial fitting for each data point. At position n, the polynomial expression is:

$$p[n] = \sum_{j=0}^{N} \alpha_j n^j \tag{1}$$

Where $p[n]$ represents the value of the fitted polynomial, i.e. the fitted data point; α_j represents the coefficients of the polynomial; N represents the order of the polynomial ($N \le 2M + 1$).

The difference between the fitted polynomial $p[n]$ and the original data $x[n]$ is measured based on the least squares metric and the solution of the polynomial is calculated:

$$\varepsilon_N = \sum_{n=-M}^{M} (p[n] - x[n])^2 \tag{2}$$

where ε_N denotes the fitting error; n is the position index; and M is the half-width of the window length.

Through testing different parameter combinations, optimal filtering effect was achieved with window width of 11 and polynomial order of 4, yielding MSE of 1.06×10^{-6} and SNR of 36.31 dB. The SG filter exhibits good performance and effectively maintains the overall trend of voltage degradation data while reducing noise.

3 Prediction Model

3.1 GRU Model

Gated recurrent units (GRU) are a variant of recurrent neural networks designed to address the gradient vanishing problem in traditional RNNs when processing long sequences. The GRU network reduces the gating signals to two [21], offering fewer parameters and a more streamlined structure compared to LSTM networks, while still maintaining strong performance in time series processing.

The training parameters for the GRU model were configured as follows: batch size was set to 4, with each batch containing a data length of 256. The GRU network structure comprised two layers, with output dimension set to 512, and rectified linear unit (ReLU) was selected as the activation function in the hidden layers. To mitigate model overfitting, several parameter combinations were tested, ultimately determining optimal model performance with an initial learning rate (LR) of 1×10^{-4} and Dropout of 0.1.

3.2 iTtansformer Model

While Transformer models have achieved remarkable success in natural language processing, they face challenges in time series prediction tasks, particularly for RUL prediction. Traditional embedding approaches target the channel dimension of features, mapping channel features to higher-dimensional space for feature decomposition. In contrast, Liu et al [22] proposed a transposed Transformer network model (iTransformer), whose embedding approach focuses on processing the sequence length dimension of mini-batches. This method maps sequence length data from each feature channel to higher dimensions, enabling subsequent feature extraction networks to focus more on temporal relationships within each feature channel.

The iTransformer network employs Transformer's Encoder with Multi-head Attention Mechanism at its core, which is formulated as:

$$Attention(Q,K,V) = Softmax\left(\frac{QK^T}{\sqrt{d_k}}\right)V \tag{3}$$

Where Q, K, V are the query, key, and value, respectively, and d_k is the vector length. When computing the output of each input, the algorithm takes into account the relationship between that input and all the elements before and after it in the sequence, and thus usually has better performance in long sequence prediction tasks.

Compared to the GRU network, the iTransformer model employs a self-attention mechanism capable of simultaneously processing information at all positions in the sequence, enabling greater parallelism during training and inference. This significantly improves efficiency and reduces computational costs. The iTransformer model encoder comprises 2 layers, with a convolutional layer output dimension of 2048 and embedding dimension of 512. The multi-head attention mechanism utilizes 8 heads. The LR and number of descending layer parameter settings remain consistent with those of the GRU network.

3.3 Hybrid Prediction Model

Since data-based prediction methods typically fail to capture internal cell degradation information and struggle to handle voltage rebound phenomena, this paper proposes a hybrid prediction model combining an impedance-voltage mathematical model with a CNN for impedance analysis. The life prediction workflow of the hybrid model is as follows:

(1) Analyze EIS to understand electrochemical degradation mechanisms and identify key parameters characterizing performance degradation.

(2) Establish ECM based on EIS analysis to reflect electrochemical behavior of each fuel cell component.

(3) Establish CNN-based impedance prediction model to accurately predict future impedance parameters.

(4) Establish linear impedance-voltage mathematical model to deduce future voltage changes.

(5) Characterize cell degradation trends using voltage variation.

EIS analysis reveals that fuel cell EIS typically contains three overlapping semicircles representing anode activation loss, cathode activation loss, and mass transfer loss. The EIS obtained in this study shows two semicircles corresponding to cathode activation loss and mass transfer loss. The semicircle characterizing anode activation loss is not observed due to overlap with cathode activation loss. In the mid-frequency range (1 kHz to 2 kHz), the arc appears slightly concave rather than a full semicircle, indicating non-ideal capacitance effects likely attributable to porous electrode structure [23].

As shown in Figure 2, Z_{ohm} consists of L_w and R_{ohm} in series to characterize the ohmic loss, Z_{act} consists of R_{act} and CPE_1 in parallel to characterize the activation loss, and Z_{conj} consists of R_{conj} and CPE_2 in parallel to characterize the mass transfer loss.

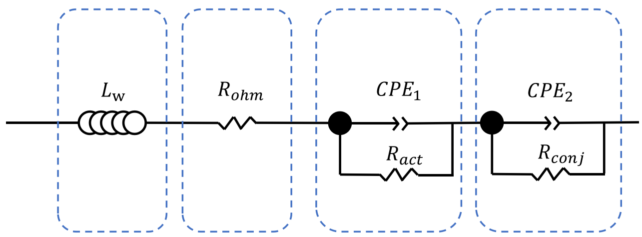


Figure 2 Equivalent circuit model of DMFC

The formulas for the full-size equivalent circuit model are specified below. The formula for ohmic loss is given below:

$$Z_{ohm} = iwL_w + R_{ohm} \tag{4}$$

where i is the imaginary number $\sqrt{-1}$; ω is the angular frequency, which is related to the regular frequency f by $w = 2\pi f$; L_w denotes the inductive resistance; and R_{ohm} denotes the ohmic resistance.

The formula for activation loss is as follows:

$$Z_{act} = Z_{CPE1} + R_{act} \tag{5}$$

The equation for mass transfer loss is as follows:

$$Z_{conj} = Z_{CPE2} + R_{conj} \tag{6}$$

Due to the porous structure of the electrodes, the capacitance C is replaced by a Constant Phase Element (CPE). The impedance expression for a CPE is usually as follows:

$$Z_{CPE} = \frac{1}{C(i \cdot \omega)^{\alpha}} \tag{7}$$

This yields the total internal loss Z of the cell as:

$$Z = Z_{ohm} + Z_{act} + Z_{conj} \tag{8}$$

Based on the developed ECM, EIS data at different time points during degradation were fitted using Zview analysis software. Table 1 shows parameter values obtained at different moments.

Table 1 Demonstration of Equivalent Circuit Model (ECM) model parameters for DMFC degradation stage

Time	u_{work}	R_{ohm}/Ω	R_{act}/Ω	R_{conj}/Ω
0h	0.381	0.45652	0.17783	4.527
120h	0.345	0.51371	0.19904	4.328
504h	0.229	0.63053	0.37384	4.252
744h	0.181	0.7631	0.44711	4.097
912h	0.137	0.82943	0.45059	4.377

Analysis reveals a clear linear relationship between output voltage and impedance. Using least squares regression, linear equations between R_{ohm}, R_{act} which are respectively related to the voltage u as:

$$R_{ohm} = -1.691u + 1.086 \tag{9}$$

$$R_{act} = -0.913u + 0.60002 \tag{10}$$

Regarding the performance evaluation indexes of the constructed linear function, they are shown in Table 2 below. The data in the table show a clear linear relationship between the data of the two impedance parameters and the voltage, and the predicted values of the model are very close to the actual observed values, which proves the accuracy of the linear model.

Table 2 Performance evaluation metrics display for linear function fitting

Parameters	R^2	ε	|Pearson coefficient|
R_{ohm}	0.96765	0.00158	0.98369
R_{act}	0.97217	0.00177	0.98599

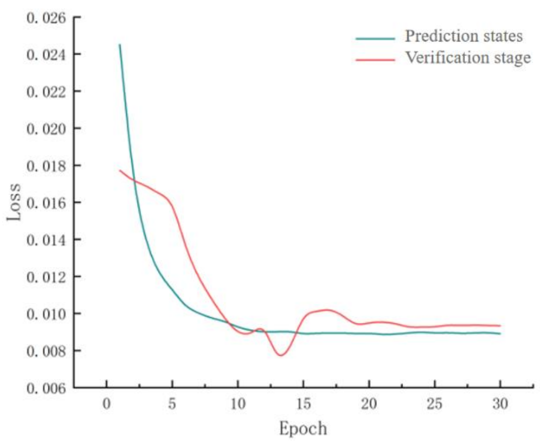
All R^2 values close to 1 indicate effective fitting of the linear relationship between impedance and voltage. Pearson coefficients close to 1 confirm strong linear correlation.

Based on impedance parameters from ECM fitting, future impedance values are predicted using CNN. The optimized CNN network primarily comprises an input layer, convolutional layer, and fully connected layer. The CNN input layer consists of seven neurons receiving time and feature column data, mapping input dimension to 64 dimensions through a linear layer. The convolutional layer extracts features along the time dimension with output dimension of 128.

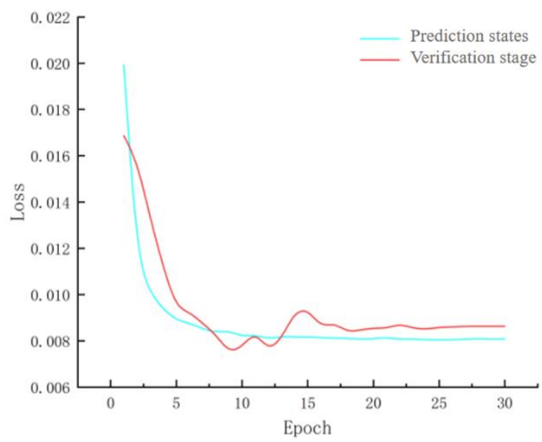
4 Results and Discussion

The sampling interval was set to 5 minutes per data point. The dataset was divided with 70% allocated for training and 30% for testing. The failure threshold was defined at 0.15 V (initial voltage 0.37 V), corresponding to end-of-life time of 769 h.

Figure 3 shows loss trends for GRU and iTransformer models during training. Both models show gradual stabilization after approximately 25 epochs.



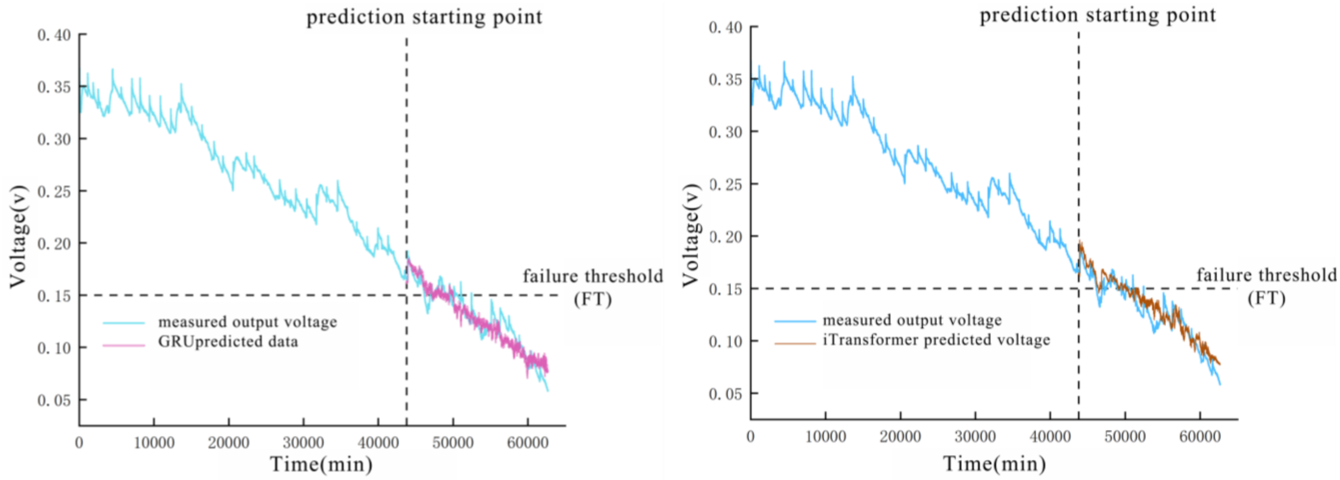
(a) Loss trends of GRU network



(b) Loss trends of iTransformer network

Figure 3 Trends in loss of different models

Figure 4 presents voltage prediction curves. The GRU model reflects overall voltage decline but proves ineffective in handling local fluctuations caused by voltage rebound. The iTransformer model not only predicts degradation trends more accurately but also provides more detailed degradation characteristics.



(a) Prediction results based on GRU network (b) Prediction results based on iTransformer network

Figure 4 Voltage prediction curves for DMFCs

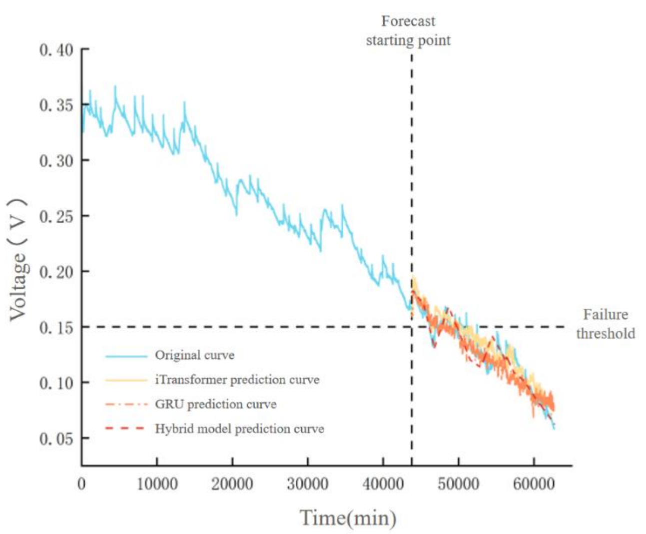


Figure 5 Plot of RUL results for different models predicting DMFC

Figure 5 shows RUL prediction results comparing GRU, iTransformer, and hybrid models. The hybrid prediction model accurately captures the voltage rebound phenomenon and effectively reflects cell performance decay.

To quantitatively evaluate prediction accuracy, RMSE, MAE, and R^2 were selected. The specific comparative evaluation metrics for the three models are detailed in Table 3. Based on the presented data, from an RMSE perspective, the iTransformer model performs optimally, followed by the hybrid prediction model, while the GRU model exhibits relatively higher RMSE, indicating comparable prediction accuracy among all three models. According to R^2 values, the hybrid prediction model slightly outperforms both iTransformer and GRU models in terms of fitting, demonstrating superior goodness-of-fit. MAE results indicate that the iTransformer model achieves the best prediction accuracy, with the hybrid prediction model ranking second, and the difference between them being minimal—both exhibiting strong prediction performance.

Table 3 Specific presentation of evaluation metrics for different models

Model evaluation index	**GRU model**	**iTransformer model**	**Hybrid prediction model**
Root mean square error	0.0117	0.0105	0.0106
Determination coefficient	0.8458	0.8737	0.8758
Mean absolute error	0.0093	0.0086	0.0084
Time error	11h	5h	1h

From RMSE perspective, the iTransformer model performs optimally, followed by the hybrid prediction model. According to R^2 values, the hybrid prediction model slightly outperforms both iTransformer and GRU models.

MAE results indicate the iTransformer model achieves best prediction accuracy, with hybrid model ranking second—both exhibiting strong performance.

Notably, time prediction errors show significant variation: GRU model exhibits 11-hour error, iTransformer achieves 5 hours, while hybrid model attains only 1 hour. This demonstrates the hybrid model's superiority in temporal prediction accuracy.

This result can be attributed to the hybrid method incorporating internal impedance parameter variation patterns, enabling more accurate capture of actual fuel cell voltage cyclic fluctuations. Both iTransformer and GRU models rely solely on historical data evolution, exhibiting limited adaptability when handling periodic voltage fluctuations.

5 Conclusion

This paper presented a hybrid prediction framework combining impedance-voltage mathematical modeling with CNN-based impedance analysis for DMFC lifetime prediction. The main contributions are:

(1) An improved iTransformer prediction network was proposed for long time series data prediction. The method employs SG filtering for noise reduction and utilizes feature importance ranking to extract effective features affecting voltage degradation. The model achieves prediction time error of only 5 hours for the complete prediction stage, indicating capability to accurately reflect real long-term voltage trends.

(2) Comprehensive validation of iTransformer prediction accuracy was conducted through three distinct performance tests. Side-by-side comparison with GRU demonstrated superior performance. Evaluation under different inference length tasks showed high prediction accuracy and strong generalization. Validation using different experimental datasets confirmed robust robustness and adaptability.

(3) To address voltage rebound phenomena, an adapted full-circuit ECM was constructed through electrochemical mechanism analysis of DMFC EIS. Impedance parameters characterizing internal state changes were obtained, with CNN employed to predict future impedance variations. By establishing a mathematical model between impedance and voltage, cell output voltage was obtained through back propagation. Results demonstrate the hybrid model effectively handles voltage rebound phenomena and more accurately captures fuel cell degradation patterns.

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