

Model-based health state estimation method for proton exchange membrane fuel cells

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Keywords: Fuel cell vehicle, State of health estimation, Particle filter

Abstract: In order to control the output power of proton exchange membrane fuel cell (PEMFC) more accurately during the aging process, the power-current curve was selected as the state of health (SOH) index. Aiming at the estimation of health status indicators, the mapping relationship between the fuel cell power and the aging of internal components was established. Based on the polarization curve, the semi mechanism power attenuation model was derived. The least square algorithm is used to fit the initialization parameters. The particle filter algorithm was employed to estimate the fuel cell SOH based on the semi mechanism power attenuation model. The experimental results show that the model estimation method based on regularized particle filter algorithm adopted in this paper can attribute to estimating the performance attenuation trend of PEMFC.

1. Introduction

The application scenarios and driving conditions of automobiles are very complicated, which requires the vehicle fuel cell system to have good durability and reliability. The durability of fuel cell system is different under different usage scenarios^[1]. The service life and durability of proton exchange membrane fuel cell (PEMFC) is a difficult problem in its commercialization process. Prediction and state of health (SOH) management technology is considered to be one of the new methods to improve the durability of fuel cells. The performance degradation process of fuel cell is a multi-phase coupled nonlinear process, and it is very difficult to model its performance degradation process. The methods of fuel cell SOH estimation are generally divided into three categories: model-based estimation method, data-driven estimation method and hybrid method-based estimation method^[2].

Aiming at the problem of fuel cell aging estimation, this paper proposes a SOH model estimation method based on particle filter (PF) algorithm based on semi-empirical-semi-mechanism power attenuation model (hereinafter referred to as semi-mechanism model). Then, simulation experiments and result analysis are carried out to compare the accuracy of the SOH estimation results of the proposed method and the existing research methods.

2. SOH estimation method

2.1 Selection of health status indicators

Different from the previous researchers who mostly use the remaining useful life as the ultimate goal to study the estimation method of the selected health status index, this paper studies the performance characterization of the output power-current curve as a health status index from the perspective of fuel cell control and convenient practical application. The purpose is to achieve more accurate control and prolong the life of PEMFC by updating the power-current curve in the controller during the aging process.

2.2 SOH estimation method based on model

2.2.1 Semi-mechanism model

There are three common aging attenuation models: mechanism model, empirical model and semi-empirical-semi-mechanism model. The semi-mechanism model is a combination of mechanism model and empirical model. Compared with the other two models, it has the advantages of small amount of data required, small amount of calculation, high calculation accuracy and no need for complex mechanism modeling.

The half-mechanism power attenuation model of fuel cell polarization curve can characterize the aging state of fuel cell through the external output characteristics of fuel cell. The output power-current formula of the fuel cell stack is as follows:

$$P = N \left[E_{ocv} - a \ln \left(\frac{I}{i_0 A} \right) - iR + b \ln \left(1 - \frac{I}{i_L A} \right) \right] I \quad (1)$$

Where P is the output power of the fuel cell stack, N is the number of single cells in the fuel cell stack, I is the output current of the fuel cell stack, A is the electrochemical active area of the single cell, and R is the internal resistance of the single cell.

The parameters in the formula (1) have little change in the exchange current density i_0 under both static and dynamic load conditions during the aging process of the fuel cell, and the change trend and amplitude of the internal resistance R and the limiting current density i_L are similar^[3].

In the existing research^[3-6], the remaining service life of fuel cell is taken as the research target, and the single aging factor α is used to characterize the aging rate of single cell internal resistance and single cell limiting current density to evaluate the remaining service life of fuel cell. In this paper, the aging factors α and β are introduced to characterize the change trend of parameters in the aging process, where α is used to characterize the aging rate of open circuit voltage, β is used to characterize the aging rate of single cell internal resistance and single cell limit current density. The variation trend of each parameter with time during the aging is as follows:

$$E_{ocv}(t) = E_{ocv}(0) \cdot (1 + \alpha(t)) \quad (2)$$

$$R(t) = R(0) \cdot (1 + \beta(t)) \quad (3)$$

$$i_L(t) = i_L(0) \cdot (1 - \beta(t)) \quad (4)$$

Where $E_{ocv}(t)$, $R(t)$ and $i_L(t)$ are the open circuit voltage, internal resistance and limiting current density of single cell at time t , respectively. $E_{ocv}(0)$, $R(0)$ and $i_L(0)$ are the open circuit voltage, internal resistance and limiting current density of the single cell at the initial time, respectively.

The parameters at the initial time can be determined by fitting the polarization curve data at the

initial time. From (1) to (4), the polarization curve formula and power-current formula of fuel cell at time t can be obtained as follows:

$$V(t) = E_{ocv}(t) - a \ln\left(\frac{I}{i_0A}\right) - iR(t) + b \ln\left(1 - \frac{I}{i_L(t)A}\right) \quad (5)$$

$$P(t) = N \left[E_{ocv}(t) - a \ln\left(\frac{I}{i_0A}\right) - iR(t) + b \ln\left(1 - \frac{I}{i_L(t)A}\right) \right] I \quad (6)$$

where $V(t)$ and $P(t)$ are respectively the voltage and power of the fuel cell stack at the moment. The formula (6) is a half-mechanism power attenuation model. After completing the estimation of the aging rate parameters α and β , the estimation results of the power-current curve can be obtained by the formula (6).

2.2.2 Particle filter algorithm

In order to avoid the phenomenon of particle degradation, resampling is carried out, that is, resampling is carried out in the discrete particle swarm with posterior probability density. After resampling, the particles in the particle swarm satisfy the characteristics of independent and identical distribution. The weight of each particle in the new particle swarm is $1/N$. The strategy of this method is to replicate the particles with larger weights in the particle swarm to replace the particles with smaller weights in the particle swarm, as shown in Figure 1. In Fig.1, the circle represents the particle, and the radius of the circle represents the weight of the particle. The Resampling behavior between $t-1$ time and t time in the graph is resampling. After obtaining the posterior probability density at $t-1$ time, resampling is performed according to the weight of the particle.

Gordon et al.^[7] first applied the resampling method to the sequential importance sampling algorithm in 1993, which is the basic model of the PF algorithm, namely the sequential importance resampling algorithm. In this algorithm, there are three steps at each moment: prediction stage, update weight stage and resampling stage. Before the prediction stage, particle swarm initialization or resampling method is needed to ensure the diversity of particles.

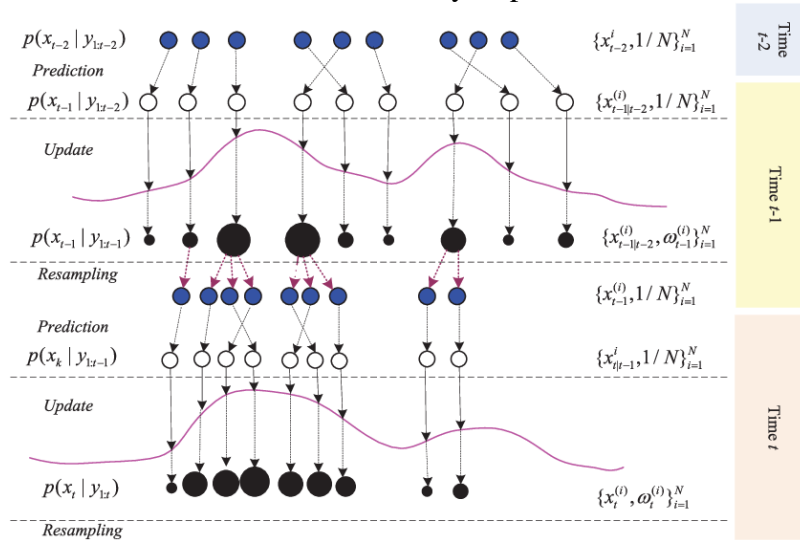


Figure 1: Resampling method

2.2.3 Model-based SOH estimation method

PF algorithm^[8] is often used to study the problem of parameter estimation based on model

aging^[6,9]. The core idea is to simulate the probability density function through a set of randomly distributed discrete samples, and use the weighted mean of the samples instead of the integral operation to obtain the optimal estimate of the system state.

The SOH estimation method based on semi-mechanism power attenuation model and PF algorithm is selected. Based on the semi-mechanism model shown in Formula (6), the aging rate α and β and their corresponding aging rate γ and δ are selected as state variables. The state space expression is established as follows:

$$\begin{cases} X_{k+1} = AX_k + W_k \\ Y_{k+1} = g(X_k, U_k) + V_k \end{cases} \quad (7)$$

where X_k is the state variable containing the aging rate and the aging rate, $X_k = [\alpha_k, \beta_k, \gamma_k, \delta_k]^T$, γ_k and δ_k are the derivatives of α_k and β_k , respectively, A is the state transition matrix, Y_k is the stack output power, W_k and V_k are the state variable noise and the observed value noise, $g(X_k, U_k)$ is the output equation, as follows:

$$A = \begin{bmatrix} 1 & \Delta t & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & \Delta t \\ 0 & 0 & 0 & 1 \end{bmatrix} \quad (8)$$

$$g(X_k, U_k) = N \left[E_{\text{ocv}}(0) \cdot (1 + \alpha(t)) - a \ln \left(\frac{I}{i_0 A} \right) - iR(0) \cdot (1 + \beta(t)) + b \ln \left(1 - \frac{I}{i_L(0) \cdot (1 - \beta(t)) A} \right) \right] I \quad (9)$$

where Δt is the time interval, which is the time interval in the data after data preprocessing in this paper.

The model-based SOH estimation method includes two parts: training phase and estimation phase. The training phase is the step of obtaining the estimated value of the state variable by the PF algorithm at a certain moment. In the training phase, the experimental test data can be used as the observation value to update the posterior estimation of the state variable. In the estimation stage, the aging rate is estimated by using the state variable results obtained in the training stage, so as to realize the estimation of power change with time.

3. Experiment and Result Analysis

3.1 Parameter fitting and data analysis method of half-mechanism power attenuation model

Table 1: Boundary conditions of parameters in semi-mechanism model parameter fitting

Parameter	Constraint condition
Single cell open circuit voltage E_{ocv} (V)	0.9~1.2
Constant coefficient a	0~1
Constant coefficient b	0~1
Single cell exchange current density i_0 (A/cm ²)	0~0.01
Single cell internal resistance R (Ω /cm ²)	0~0.6
Single cell limit current density i_L (A/cm ²)	0~1.5

The nonlinear least squares algorithm based on Levenberg-Marquardt optimization algorithm^[10] is selected to fit the parameters of formula (1) in the semi-mechanism model with the objective of minimizing the sum of squares of the fitting voltage V and the measurement voltage V error. The constraint conditions of the parameters^[11] are shown in Table 1, and the results of the fitting

parameters are shown in Table 2. Some of the parameters in the results correspond to $E_{ocv}(0)$, $R(0)$ and $i_L(0)$ in formulas (2) ~ (4).

Table 2: Fitting results of semi-mechanism aging model parameters

Parameter	Fitting result
Single cell open circuit voltage E_{ocv} (V)	0.9982
Constant coefficient a	1.012×10^{-4}
Constant coefficient b	1.698×10^{-4}
Single cell exchange current density i_0 (A/cm ²)	1.010×10^{-3}
Single cell internal resistance R (Ω /cm ²)	0.084
Single cell limit current density i_L (A/cm ²)	1.30

In the PF algorithm, the observation estimate can characterize the accuracy of the state variable estimate. Considering that the trend of power changing with time is more intuitive, the power estimation results can be used to show the accuracy of the state variable estimation results. In the presence of observations, the PF algorithm can accurately estimate the state variables. Therefore, the PF algorithm can accurately estimate the aging rate at all times in the test experiment. The experiment does not measure the polarization curve and aging rate at all times. The aging rate estimation results obtained by this method can be used as the aging rate reference value for error comparison analysis and verification of the effectiveness of the estimation method in this paper.

The SOH estimation method based on the model is used to estimate the aging rate. The power-current curve estimation results can be obtained by using the aging rate estimation results combined with Formula (6). The number of particles in the PF algorithm is set to 1000, the noise variance of α is 0.001, the noise variance of β is 0.002, the noise variance of γ and δ is 0.0005, and the noise variance of the observed value is 1.0. The Root Mean Square Error (RMSE) is usually used to evaluate the accuracy between the estimated results and the experimental data. The formula is as follows:

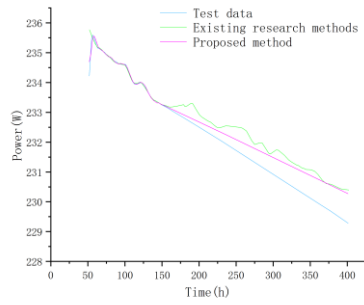
$$\text{RMSE} = \sqrt{\frac{1}{n} \sum_{i=1}^n (\hat{y}_i - y_i)^2} \quad (10)$$

where \hat{y}_i represents the estimated value of the estimation method, y_i represents the real value in the test data result, and n is the number of estimated values.

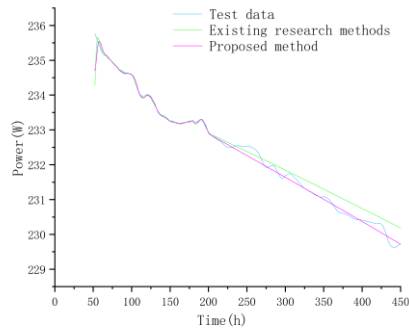
3.2 Comparison between the proposed method and the existing research methods

The semi-mechanism model based on the polarization curve has been used for aging research, but they use a single aging rate from the perspective of estimating the remaining useful life. They only consider the aging of single cell internal resistance and limiting current density in formula (1), and do not consider the aging of single cell open circuit voltage. This paper considers the changes of single cell open circuit voltage, internal resistance and limiting current density from the perspective of estimating the power-current curve. Compared with the existing research methods, the research method in this paper considers the changes of more parameters in the aging process, and the estimation results of power should be more accurate.

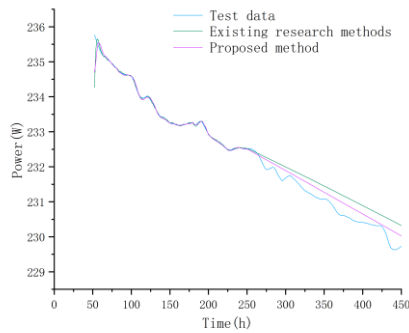
In this paper, four groups of experiments are carried out to compare the differences in power estimation results between the existing research methods and the research methods in this paper. The length of training time in the four groups of experiments is different, and the length of estimation time is the same, which is 250 hours. The length of training time in experiments 1, 2, 3 and 4 is 100, 150, 200 and 250 hours, respectively. The experimental results correspond to a, b, c and d in Fig.3. The corresponding RMSE results for each experiment are shown in Table 3.



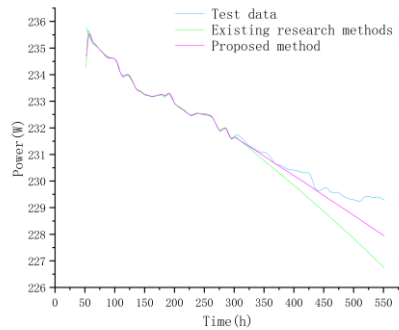
a) The training time is 100 hours and the estimated time is 250 hours.



b) The training time is 150 hours and the estimated time is 250 hours.



c) The training time is 200 hours and the estimated time is 250 hours.



d) The training time is 250 hours and the estimated time is 250 hours.

Figure 2: Comparison of power estimation results between the proposed method and the existing research methods

Figure 2 shows the comparison between the training results, estimation results of the existing research methods and the training results, estimation results and test data results of the research methods in this paper. It can be seen from the figure that the estimation results of the power in the training stage of the four experiments are more accurate, and the linear trend of power attenuation can be accurately estimated in the estimation stage. However, the proposed method has higher estimation accuracy than the existing research methods. Compared with the test data results, the estimation results of the existing research methods gradually deviate from the test data results with the increase of the estimation time, and the estimation results may be higher or lower than the test results. The estimation results of this method can more accurately fit the test results, and the relative error is smaller. It can be seen from Table 3 that under four different training durations, compared with the estimation results of the existing research methods, the estimation results of this method have smaller errors in both the training stage and the estimation stage. The RMSE decline rate in the training stage is 7 % -18 %, and the RMSE decline rate in the estimation results is 45 % -65 %.

Table 0: The RMSE of power estimation and test data results of the estimation method in the existing research and this study

Experiment		1	2	3	4
Training stage	Existing research	0.195	0.146	0.143	0.125
	This study	0.167	0.135	0.118	0.104
	Error reduction rate	14.36%	7.53%	17.48%	16.80%
Estimation stage	Existing research	0.717	0.281	0.455	1.149
	This study	0.246	0.127	0.247	0.537
	Error reduction rate	65.69%	54.80%	45.71%	53.26%

3.3 Comparison of power-current curve estimation results

In this paper, the power-current curve estimation results are obtained by using the aging rate estimation results under single current and Formula (6). Figure 3 shows the reference results of power-current curves for 100,300,500 and 700 hours. It can be seen from the figure that the power-current curve decreases with time during the aging process. When the current is relatively small, the degree of power decline is not very obvious, and when the current is large, the degree of power decline is very obvious. Comparing the power-current curves of 100 and 700 hours, the root mean square error of power is 1.64W in the range of 0-50A, and in the range of 50-100A, the root mean square error of power is 21.10W, that is, the degree of power decline in the large current range is much larger than that in the small current range. This is because the absolute value of power in the small current interval is relatively small, and the absolute value of power in the large current interval is relatively large. Therefore, in order to more intuitively display the estimation results of the power-current curve, only the power-current curve in the 50-100A interval is displayed.

The power-current curve estimation results obtained by the aging rate reference value are compared with the power-current curve results measured in the test experiment. The results are shown in Figure 4. It can be seen that the trend of the power-current curve estimation results and the experimental test results is completely consistent, and the accuracy rate is as high as 99.85 %. The results of 24-36A and 64-76A are enlarged respectively, corresponding to two different current intervals. The numerical error in the two current intervals shown in the enlarged figure is very small, which is 0.06W. There is a big gap only in the current range (92-100A in the figure), and the RMSE is 0.46W.

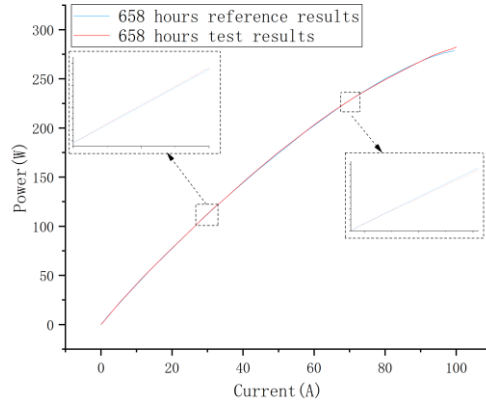


Figure 3: Comparison of reference power-current curves at different moments

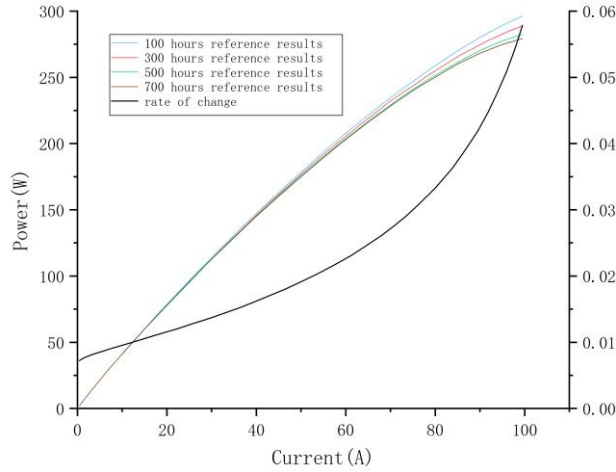


Figure 4: Comparison between power-current curve estimation results and experimental results

In order to intuitively show the accuracy of different methods in SOH estimation in the experiment, the aging rate estimation results and reference aging rate of the proposed method and the existing research methods in four experiments are compared. The error of power estimation value and reference power value of different methods in different current intervals is selected for analysis. Taking five fixed currents of 10A, 30A, 50A, 70A and 90A as examples, the RMSE error results are shown in table 4.

The experimental results show that the accuracy of the SOH estimation results of this method is 99 %, which is higher than the estimation results of the existing research methods. Among them, the RMSE reduction rate in the small current range is about 90 %, and the RMSE reduction rate in different experiments in the medium and high current ranges is different, and the overall is between 10 % and 90 %. Compared with the existing research methods that only consider the changes of the two parameters of internal resistance and limiting current density, the adopted model considers the changes of the three parameters of single cell open circuit voltage, internal resistance and limiting current density during the aging process. Therefore, the semi-mechanism power attenuation model in this paper can more accurately estimate the power change trend and power-current curve during the aging process.

Table 4: The error of power estimation and reference value in experiments 1-4

Experiment		10A	30A	50A	70A	90A
1	Existing research	0.188	0.436	0.452	0.088	0.124
	This study	0.015	0.043	0.068	0.082	0.086
	Error reduction rate	92.02%	90.14%	84.96%	6.82%	30.65%
2	Existing research	0.236	0.376	1.057	1.340	1.383
	This study	0.013	0.039	0.063	0.084	0.099
	Error reduction rate	94.49%	89.63%	94.04%	93.73%	92.84%
3	Existing research	0.269	0.772	1.214	1.551	1.625
	This study	0.005	0.033	0.092	0.202	0.437
	Error reduction rate	98.14%	95.73%	92.42%	86.98%	73.11%
4	Existing research	0.264	0.553	0.412	0.502	3.551
	This study	0.055	0.189	0.366	0.439	1.059
	Error reduction rate	79.17%	65.82%	11.17%	12.55%	70.18%

4. Conclusion

In this paper, the power-current curve is selected as the health state index from the perspective of vehicle control. Based on the updated polarization curve in the process of performance degradation, a semi-mechanism power aging model is proposed. Based on the previous research, the open circuit voltage is considered. The change in the aging process increases the number of aging factors in the aging model to achieve more accurate estimation results. Then, according to the nonlinear characteristics of fuel cell performance attenuation, the PF algorithm is selected to estimate the SOH, and the aging rate estimation model is designed based on the PF algorithm, and the power-current curve is estimated by combining the power attenuation model. Simulation experiments are carried out on the test data set and compared with the experimental test data. The results show that the model method can predict the long-term performance degradation trend. Compared with the existing research methods, the research method in this paper can more accurately estimate the power-current curve during the aging process through the aging rate reference value and the power attenuation model.

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