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Estimating the Remaining Useful Life of Proton Exchange Membrane Fuel Cells under Variable Loading Conditions Online

Penghao Wang [1,2], Hao Liu [3], Ming Hou [1], Limin Zheng [1], Yue Yang [1,2], Jiangtao Geng [1], Wei Song [1,*] and Zhigang Shao [1,*]

1 Dalian National Laboratory for Clean Energy, Dalian Institute of Chemical Physics, Chinese Academy of Sciences, Dalian 116023, China; wangpenghao@dicp.ac.cn (P.W.); houming@dicp.ac.cn (M.H.); zhenglimin@dicp.ac.cn (L.Z.); yue.y@outlook.com (Y.Y.); gengjt@dicp.ac.cn (J.G.)
2 University of Chinese Academy of Sciences, Beijing 100049, China
3 State Key Laboratory of Industrial Control Technology, College of Control Science and Engineering, Zhejiang University, Hangzhou 310027, China; haoliu7850052@zju.edu.cn
* Correspondence: songwei05@dicp.ac.cn (W.S.); zhgshao@dicp.ac.cn (Z.S.)

Abstract: The major challenges for the commercialization of proton exchange membrane fuel cells (PEMFCs) are durability and cost. Prognostics and health management technology enable appropriate decisions and maintenance measures by estimating the current state of health and predicting the degradation trend, which can help extend the life and reduce the maintenance costs of PEMFCs. This paper proposes an online model-based prognostics method to estimate the degradation trend and the remaining useful life of PEMFCs. A non-linear empirical degradation model is proposed based on an aging test, then three degradation state variables, including degradation degree, degradation speed and degradation acceleration, can be estimated online by the particle filter algorithm to predict the degradation trend and remaining useful life. Moreover, a new health indicator is proposed to replace the actual variable loading conditions with the simulated constant loading conditions. Test results using actual aging data show that the proposed method is suitable for online remaining useful life estimation under variable loading conditions. In addition, the proposed prognostics method, which considers the activation loss and the ohmic loss to be the main factors leading to the voltage degradation of PEMFCs, can predict the degradation trend and remaining useful life at variable degradation accelerations.

Keywords: proton exchange membrane fuel cells; prognostics; remaining useful life; health indicator; particle filter

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1. Introduction

As fossil energy consumption continues to increase and the environment continues to deteriorate, there is an urgent need to find clean renewable energy and conversion devices. Proton exchange membrane fuel cells (PEMFCs) can directly convert chemical energy into electrical energy, the unique characteristics, such as high efficiency, high power density, no pollution, and low operating temperature, make PEMFCs be one of the most promising candidates for power generation. Therefore, PEMFCs have been used in many fields [1–3]. However, low durability and high cost hinder the commercialization process [4,5].

Although PEMFCs will inevitably exhibit performance degradation with the increase of running time, the degradation rate can be effectively slowed down by effective prognostics and health management (PHM) technology [6]. As shown in Figure 1, PHM consists of seven layers [7]: data acquisition, data processing, condition assessment, diagnostics, prognostics, decision support and human–machine interface. It aims at utilizing the real-time monitoring data of the target system to diagnose and predict its health status.

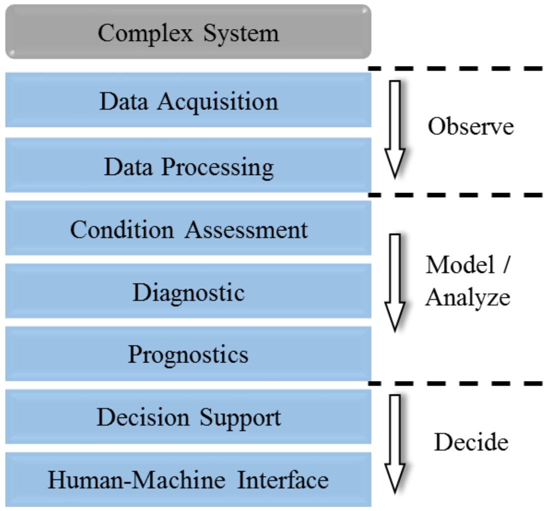

Figure 1. Prognostics and health management (PHM) architecture.

Meanwhile, once a fault is found, the PHM technology can provide alternative solutions and implement them at the right time to extend the service life of the system. Then, the maintenance strategy can be changed from "fail to fix" to "predict to prevent", which improves the safety and availability of the target system. Studies have shown that the application of PHM technology is of great significance to the durability, reliability and maintainability of PEMFCs [8], while reducing the maintenance cost [9]. This paper is focused on the prognosis layer, which aims at predicting the degradation trend and remaining useful life (*RUL*) of the target system based on the health indicators extracted from the measured data.

According to the definition of the International Organization for Standardization [10], prognostics is the estimation of time to failure and risk for one or more existing and future modes. The *RUL* is defined as the time between the predicted time (t_{pred}) and the end-of-life (EOL) time (t_{EOL}) [11].

$$RUL\left(t_{\text{pred}}\right) = t_{\text{EOL}} - t_{\text{pred}} \tag{1}$$

For PEMFCs, the US Department of Energy considers that the EOL is reached when its initial performance declines by 10% [12]. Since voltage is an effective degradation indicator of PEMFCs, this paper considers that EOL is reached when the voltage of PEMFCs drops to 90% of the initial value.

Prognostics studies of PEMFCs have made great progress in recent years, the prognostics methods can be divided into three categories: model-based methods, data-based methods and hybrid methods [8,13]. The model-based method predicts the system's degradation information based on the empirical or physical models of fuel cells, and the predictive algorithm used to be extended Kalman filter (EKF), unscented Kalman filter (UKF) and particle filter (PF) [14–16]. A detailed prediction process by PF is presented in [14], the authors adopt three different empirical models: linear model, logarithmic model and exponential model to predict the degradation trend of voltage, and the results show that the logarithmic model is more efficient, but it cannot be used under variable loading conditions. A precise degradation model is hard to build, so the data-based method is more and more popular. It predicts the degradation trend by kinds of machine learning algorithms: wavelet-based approach [6], echo state network [17,18], adaptive neuro-fuzzy inference systems [19], relevance vector machine [20,21] and so on. But a large number of high-quality data is essential. The hybrid method combines the model-based method with the data-based method to achieve complementary advantages [8]. Liu et al. [22] firstly adopt an automatic machine learning algorithm to predict the degradation trend, and then an adaptive unscented Kalman filter (AUKF) is used to estimate the RUL.

Apart from choosing a proper prognostics method, a suitable health indicator of fuel cells during the degradation process is also difficult to choose. The common health indicators in the literature are measured voltage [1,14,17,23,24], measured power [6,25,26], ECSA [27,28], model parameters [11,15,29,30] and so on. But the measured voltage and power are just suitable for constant loading conditions. Although ECSA can be used under variable loading conditions, it is difficult to measure online. The model parameters can be influenced by different materials, synthesis processes and assembly technologies. In sum, choosing a reasonable health indicator is essential and particularly important, especially under variable loading conditions. A new health indicator is proposed in this paper to overcome the shortcomings above, it can be used not only for constant loading conditions, but also for variable loading conditions online (Section 4.2).

The remaining content of this paper is organized as follows. The aging test of the PEMFCs stack is described in Section 2. Then, a model-based prognostics method including an empirical degradation model and particle filter algorithm is introduced in Section 3. Next, the prediction results including the estimated voltage, new health indicator and remaining useful life are analyzed and discussed in Section 4. Finally, the conclusions are presented in Section 5.

2. Proton Exchange Membrane Fuel Cells (PEMFCs) Stack Aging Test

A homemade PEMFCs stack consisted of 8 cells was used in the aging test, and each cell had an active area of 270 cm^2. The PEMFCs stack was composed of metal bipolar plates with parallel flow filed and MEA consisted of commercial Pt/C catalyst and Nafion membrane. The detailed information is listed in Table 1.

Table 1. Proton exchange membrane fuel cells (PEMFCs) stack components.

Components	Information
PEMFCs stack	8 cells
Active area (cm^2)	270
Anode platinum loading (mg cm^{-2})	0.2
Cathode platinum loading (mg cm^{-2})	0.4
Proton exchange membrane	Nafion®211
Flow channel	Parallel flow field

A homemade test bench was used in the aging test, the current density was controlled by the electronic load of KIKUSUI PLZ2004WB, the temperature of PEMFCs stack was controlled by the recirculating water bath, the gas flowrate and pressure was adjusted by mass flow controllers and gas regulator valves, the gas pressure was displayed by pressure gauge, the reaction gasses were humidified by bubbling method, there were two independent boilers for reaction gasses, the air humidifier was heated to the requested relative humidity, while the hydrogen humidifier was always kept at room temperature with dry hydrogen, the details about operating parameters during the aging test are summarized in Table 2.

Table 2. Operation parameters during the aging test.

Parameters	Range
Current density (A cm^{-2})	0.8~1.0
Temperature (°C)	62~67
Anode inlet pressure (bar)	0.45~0.70
Cathode inlet pressure (bar)	0.39~0.58
Anode relative humidity (%)	dry gas
Cathode relative humidity (%)	11~40

The aging test was carried out on working days and ran for about 7~9 h every day. Each time the PEMFC stack was started, the current density gradually increased from 0 to the required current density, and then gradually decreased to 0 at the end, which caused a lot of fluctuations in the raw data (Figure 2). The loading current density was mainly 1.0 A cm^{-2} during 0~620 h, and then mainly 0.8 A cm^{-2}. The sampling frequency of the raw data was 1 Hz during the aging test. However, we would usually consider the degradation phenomenon in hours or even in days in the practical engineering situation [31]. With reference to that, the raw data were resampled with an hour and de-noised. In addition, a series of polarization curves were tested at 20 h, 155 h, 237 h, 349 h, 450 h, 555 h and 646 h respectively in the aging test (Figure 3 solid lines).

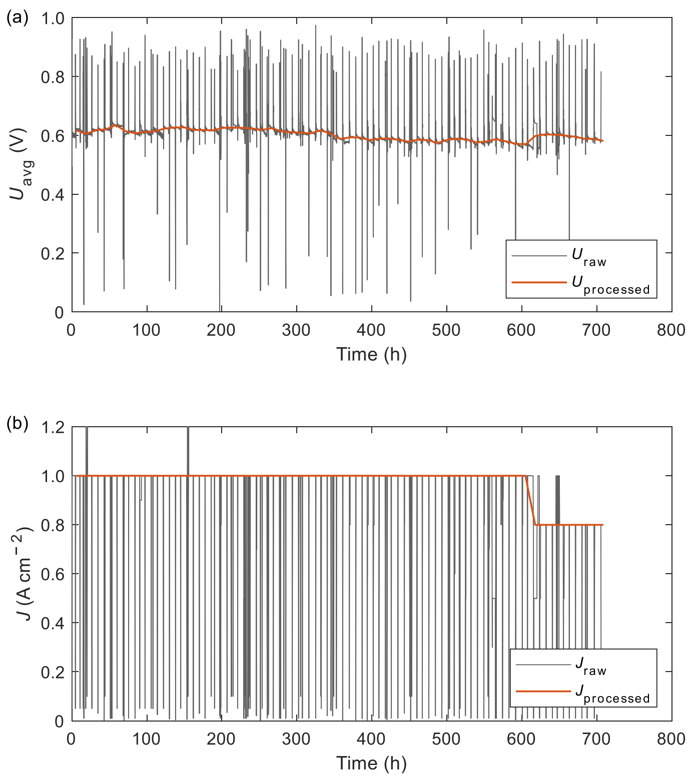

Figure 2. Average voltage (**a**) and current density (**b**) of PEMFCs stack over time.

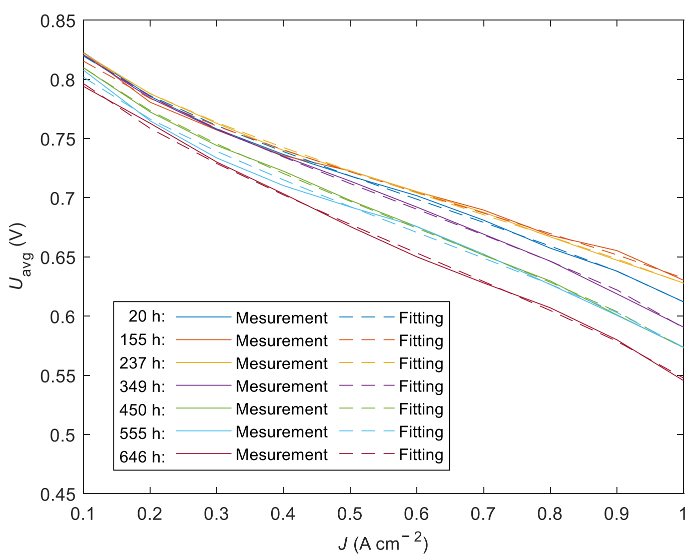

Figure 3. Polarization curves during the aging test.

3. Model-Based Prognostics Method

The total procedure of the model-based method is listed in Figure 4. An empirical degradation model of the PEMFCs stack is built in Section 3.1 based on the polarization curves. Then, the PF algorithm is introduced to estimate the degradation status in Section 3.2.

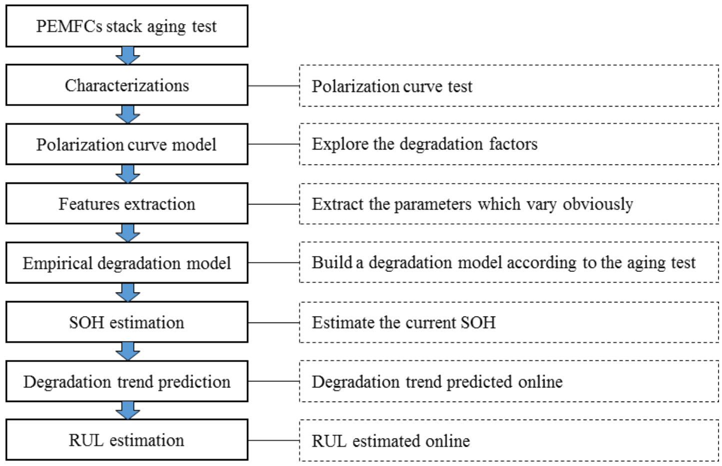

Figure 4. Model-based prognostics method for PEMFCs.

3.1. Empirical Degradation Model

To further study the degradation behavior of the PEMFCs stack, a polarization curve model proposed by Bressel et al. [15] and Blal et al. [32] was selected to fit the polarization curves at different aging stages.

$$U_{avg} = \frac{U_{stack}}{N} = E_0 - \frac{\mathcal{R}T}{n\alpha F}\ln\left(\frac{j}{j_0}\right) - jR - \left(-\frac{\mathcal{R}T}{nF}\ln\left(1-\frac{j}{j_L}\right)\right) \quad (2)$$

where U_{avg} is the average stack voltage, U_{stack} is the stack voltage, N is the number of single cell, E_0 is the open circuit voltage, T is cell temperature, j is current density, j_0 is the exchange current density, R is the total resistance and j_L is the limited diffusion current density.

Among these model parameters, only the E_0, j_0, R and j_L need to be fitted. The nonlinear Levenberg–Marquardt method is used to identify the model parameters. However, we could only obtain the local optimal solution through this method, therefore uniform initial values and acceptable fitting error are used to overcome the weakness in this paper [15]. The fitted polarization curves are shown in Figure 3 (dashed lines). It can be seen from Figure 3 that the simulation data are highly consistent with the experimental data, the average error (RMSE) is only 0.0022 V, which means the fitted model parameters are reasonable. Then the evolution of the model parameters with time is shown in Figure 5. This shows that the E_0 and j_L display no marked changes over time, so they are assumed to be constant in this paper. In contrast, the j_0 and R change significantly (decreases/increases about 40%) during the aging test, which may be caused by fuel starvation and the hydrogen–air interface under frequent start-stop conditions [33,34]. Moreover, the evolution trend of the j_0 and R seems to be a quadratic function, so the quadratic function is used to build an empirical degradation model.

Herein, we define the $\alpha(t)$ as the degradation degree at time t, so the $j_0(t)$ and $R(t)$ can be written as follows:

$$j_0(t) = j_{0,0}\cdot(1-\alpha(t)) \quad (3)$$

$$R(t) = R_0\cdot(1+\alpha(t)) \quad (4)$$

According to the empirical degradation model, the $\alpha(t)$ seems to be a quadratic function:

$$\alpha(t) = \alpha_0 + v \cdot t + \frac{1}{2} a \cdot t^2 \tag{5}$$

where α_0 is the initial degradation degree, v is the degradation speed, and a is the degradation acceleration.

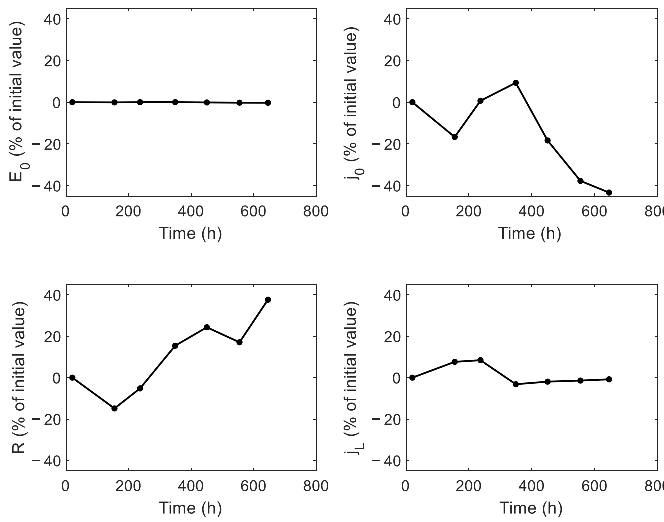

Figure 5. Evolution of the model parameters.

3.2. *Particle Filter Algorithm*

The PF is an approximate Bayesian filtering algorithm based on Monte Carlo simulation, which could handle arbitrary distributions of noises and nonlinearities theoretically [35]. The PF mainly includes three steps: prediction, update and re-sampling, the detailed framework can be seen in the references [14,26]. In this paper, PF is adopted to estimate the health status of the PEMFCs stack. The PEMFCs system can be described by the following non-linear system:

The equation of state:

$$X_k = AX_{k-1} + \omega_{k-1}, \tag{6}$$

The equation of observation:

$$Z_k = f(X_k, j_k) + \varphi_k, \tag{7}$$

where the X_k is the state of system at time k, Z_k is the average voltage of system at time k, j_k is the current density, ω_{k-1} and φ_k are Gaussian noises with variances Q and R, respectively.

$$X_k = [\alpha_k \; v_k \; a_k]^T, \tag{8}$$

$$A = \begin{bmatrix} 1 & \Delta T & 0.5\Delta T^2 \\ 0 & 1 & \Delta T \\ 0 & 0 & 1 \end{bmatrix}, \tag{9}$$

$$f(X_k, j_k) = E_0 - \frac{\mathcal{R}T}{n\alpha F} \ln\left(\frac{j_k}{j_{0,0} \cdot (1 - \alpha_k)}\right) - j_k \cdot R_0 (1 + \alpha_k) - \left(-\frac{\mathcal{R}T}{nF} \ln\left(1 - \frac{j_k}{j_L}\right)\right), \tag{10}$$

The MATLAB software is used to solve the problem. The parameters should be initialized before running the algorithm. Obviously, the $\alpha_0 = 0$ in the beginning, but it is hard to determine the degradation speed v_0 and the degradation acceleration a_0. Taking the generalization of this model into consideration, they are set uniformly to 0, hence the initial state can be written as $X_0 = [0\ 0\ 0]^T$. In order to make the program converge as soon as possible and insensible to the noises, the Q and R are chosen as [15]:

$$Q = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 10^{-12} \end{bmatrix}, \ R = 10^{-3} \tag{11}$$

4. Results and Discussion

4.1. Health Status Estimation

The health status of PEMFCs, such as degradation degree α, degradation speed v and degradation acceleration a, can be estimated online by PF by inputting the average voltage and current density of the PEMFCs stack, the average time taken for each health status estimation is only 0.13 s, and the results are shown in Figure 6. Figure 6a shows that the evolution of degradation degree α can be divided into two stages. In the first stage from 0 to 240 h, the α shows a decreasing trend, which may be caused by the activation process of PEMFCs or more favorable working conditions (e.g., the humidity or pressure of the reaction gas.), matching with that the voltage increases gradually during this stage (Figure 7 black line). In the second stage from 240 to 706 h, the α increases gradually, which corresponds to the degradation process of PEMFCs and leads to the voltage decreases little by little. It should be noted that at 620 h, the current density changed from 1.0 A cm^{-2} to 0.8 A cm^{-2}, which caused a sudden increase in voltage. Then according to Equation (10), the estimated voltage can be calculated based on the α_k and j_k, the results are shown in Figure 7 (red line), the total error (RMSE) between the real voltage and the estimated voltage is 0.0093 V, which means the estimated α is acceptable.

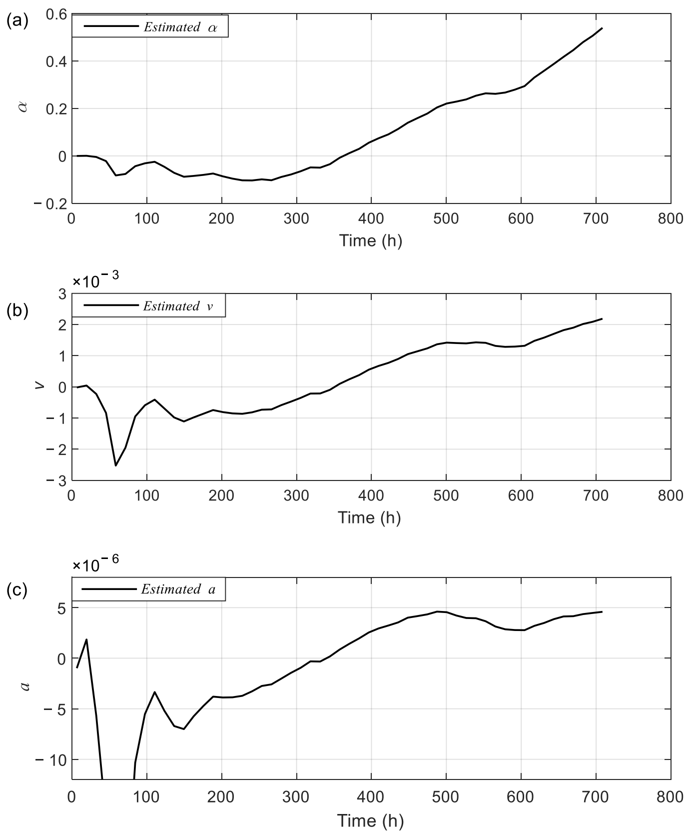

Figure 6. Degradation degree α (**a**), degradation speed v (**b**) and degradation acceleration a (**c**) estimation.

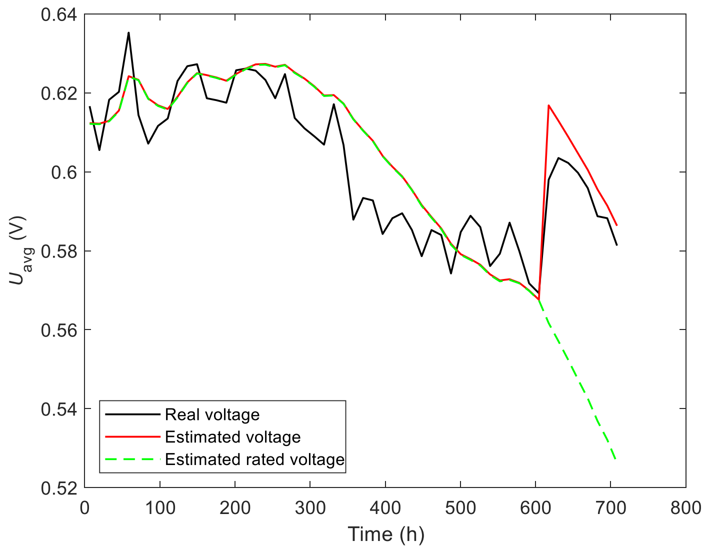

Figure 7. Voltage estimation.

Taking a close look at Figure 6a,c, it can be seen that the degradation acceleration a has an important influence on the degradation degree α. During the period of 500~600 h, a decreases rapidly, which led to the slow growth of α, and then slows down the degradation of the PEMFCs stack. In contrast, the a increases quickly after 600 h, which leads to a faster growth of α, and which means the degradation of PEMFCs is accelerated. In summary, the proposed prognostics method can predict the degradation trend at variable degradation accelerations, so it is more in line with practical demands. However, we initialized X_0 with inaccurate values, namely the $v_0 \neq 0$ and $a_0 \neq 0$ at the beginning, which causes some huge fluctuations in the early stage of v and a.

4.2. A New Health Indicator

According to the discussions in Section 4.1, although the parameter α can reflect the degradation degree of PEMFCs quite well, the precise threshold $\alpha_{\max}$ cannot be obtained beforehand. Moreover, different materials, synthesis processes and assembly technologies also have impacts on it. Hence, a more suitable health indicator should be selected to replace it. As mentioned in Section 1, although there are many health indicators (e.g., measured voltage, measured power, ECSA, model parameters et al.) that have been used in this field, few of them are applicable for variable loading conditions online. Here, we propose a new health indicator, the rated voltage, which is suitable for constant and variable loading conditions and also can be obtained online. DOE defines that the EOL is reached when PEMFC's initial performance degrades by 10%. Taking the most easily measured voltage as an example, this criterion can be realized easily under constant loading conditions, just comparing the measured voltage value with the initial value. However, the current density varies with actual demands under variable loading conditions and, correspondingly, the output voltage will change with it, which means that the measured voltage and the initial voltage value cannot be compared directly. In order to overcome this problem, the following strategy has been followed: replace the real measured voltage with the rated voltage. The detailed definition is as follows:

The real measured voltage at time k is defined as: U_{α_k,j_k}

The rated voltage at time k is defined as: $U_{\alpha_k,j_{\text{rated}}}$

Where k is the current time, α_k is the degradation degree of the PEMFCs at time k, j_k is the actual loading current density at time k, j_{rated} is the rated current density, which is a consist value. Theoretically, the rated current density can be any value, and 1.0 A cm^{-2} is chosen in this paper.

Comparing U_{α_k,j_k} and $U_{\alpha_k,j_{\text{rated}}}$, we can find that the real measured voltage U_{α_k,j_k} is not only affected by the degradation degree α_k, but also by loading current density j_k. However, the rated voltage $U_{\alpha_k,j_{\text{rated}}}$ is only affected by α_k. In this way, the new health indicator, rated voltage $U_{\alpha_k,j_{\text{rated}}}$, can successfully eliminate the influence of current density and highlight the influence of degradation degree. The estimated rated voltage $U_{\alpha_k,j_{\text{rated}}}$ at different times can be calculated according to the Equation (10) ($U_{\alpha_k,j_{\text{rated}}} = f(X_k, j_k)$) and α_k. The results are shown in Figure 7 (green line). Between 0 and 620 h, the estimated $U_{\alpha_k,j_{\text{rated}}}$ is equal to the U_{α_k,j_k}, because $j_k = j_{\text{rated}} = 1.0$ A cm^{-2}. After 620 h, the measured voltage and estimated voltage by particle filter increase rapidly because of the change of current density from 1.0 A cm^{-2} to 0.8 A cm^{-2}, while the estimated rated voltage $U_{\alpha_k,j_{\text{rated}}}$ keeps the degradation trend regardless of the changes of current density. This shows $U_{\alpha_k,j_{\text{rated}}}$ can eliminate the influence of current density and reflect the health status of PEMFCs. The polarization curve test shows that the measured voltage under 1.0 A cm^{-2} is 0.5454 V at 646 h, it is close to the estimated rated voltage 0.5530 V at the same time, which means the rated voltage $U_{\alpha_k,j_{\text{rated}}}$ is a valid health indicator for constant and variable loading conditions.

4.3. Degradation Trend Prediction

The state $X_k = [\alpha_k\ v_k\ a_k]^T$ of the PEMFCs stack at the current time k can be gotten in Section 4.1. It is worth noting that the current time is k, so the information after k is unknown, therefore the future degradation trend should be predicted based on the information before time k. Here the state X_k is used to predict the future degradation trend, and the algorithm is presented in Figure 8.

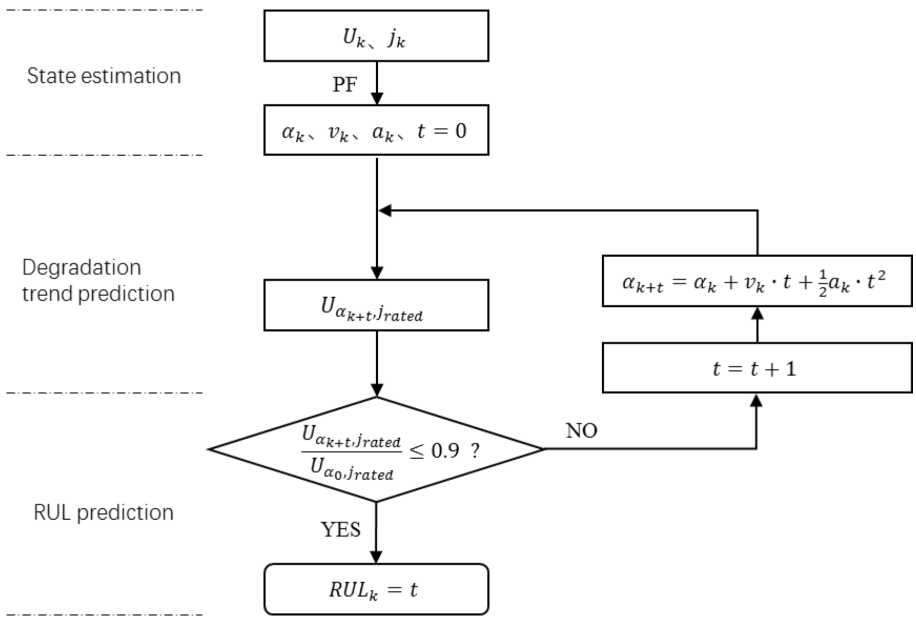

Figure 8. The degradation trend and remaining useful life (RUL) prediction algorithm.

As shown in Figure 8, in the state estimation part, the degradation degree α_k, the degradation speed v_k and the degradation acceleration a_k can be estimated by particle filter at time k. Next, in the part of degradation trend prediction, iterating the empirical degradation model (Equation (5)), the α_{k+t} can be gotten when a_k has converged. Last, the rated voltage $U_{\alpha_{k+t},j_{\text{rated}}}$ can be calculated by the predicted α_{k+t} according to Equation (10). It should be noted that the degradation trend can be predicted at every moment. For example, when the current time is 450 h, the degradation degree $\alpha_{450\text{h}}$, the degradation speed $v_{450\text{h}}$ and the degradation acceleration $a_{450\text{h}}$ can be estimated by particle filter online, then the future degradation trend can be predicted based on this information. Figure 9 shows that the predicted rated voltage is close to the estimated rated voltage, the error is only 0.0045 V, which means that the model has a good ability to predict the degradation trend.

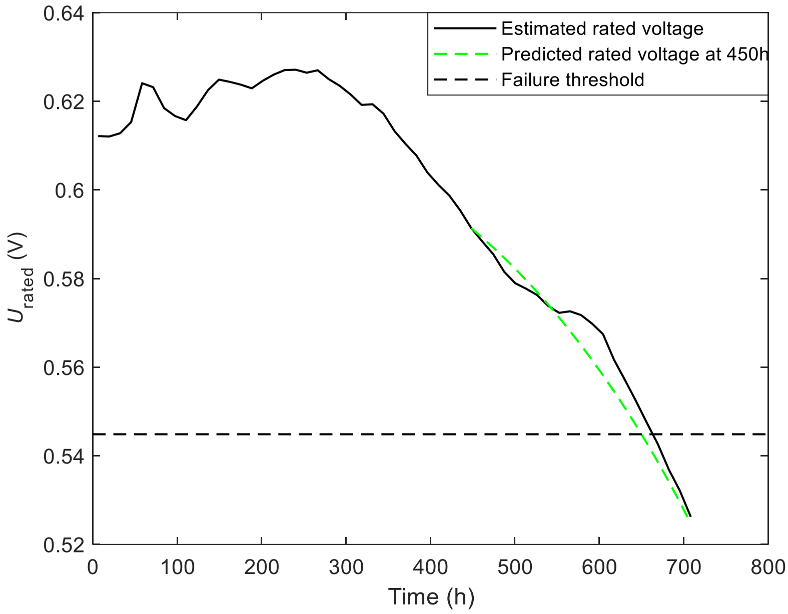

Figure 9. The degradation trend predicted at different time.

4.4. Remaining Useful Life Estimation

Following Section 4.3, the future rated voltage $U_{\alpha_{k+t},j_{\text{rated}}}$ can be estimated according to the algorithm shown in Figure 8. We repeat the degradation trend prediction part, until the $U_{\alpha_{k+t},j_{\text{rated}}}$ comes up to 90% of the initial rated voltage $U_{\alpha_0,j_{\text{rated}}}$. Then according to Equation (1), we can conclude that the time t is the remaining useful life RUL_k at time k, and the estimated RUL results at different times are shown in Figure 10. It takes about 370 h for the algorithm to converge. When a_k converges, the algorithm can successfully predict the RUL of PEMFCs. After 400 h in particular, the predicted RUL falls within the 90% confidence interval, in other words, the predicted RUL is within the bounds ±10% of the maximum lifetime, which means that the prediction algorithm has a high accuracy.

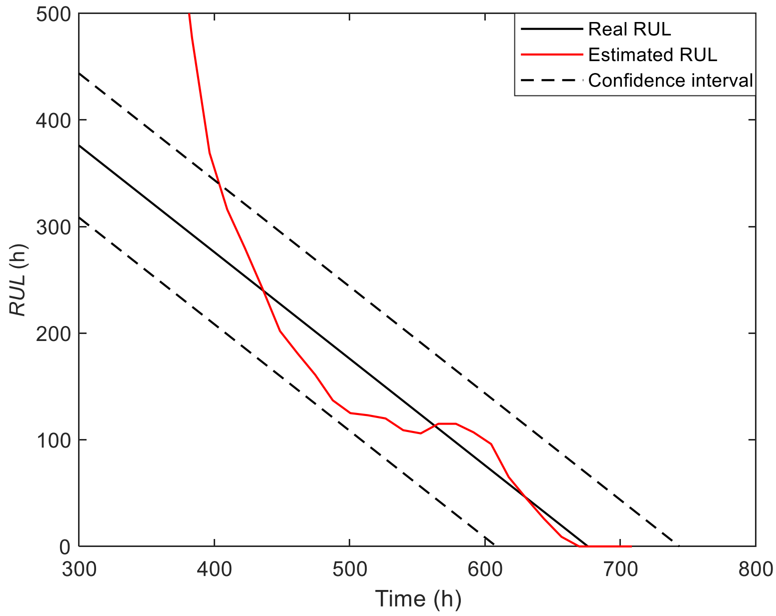

Figure 10. RUL estimation.

5. Conclusions

A PEMFCs stack aging test was carried out in our laboratory, and seven polarization curves were measured during the experiment. In order to explore the degradation reasons, a polarization curve model was used to fit the polarization curves. It found that the exchange current density j_0 and resistance R changed obviously, which may have been caused by fuel starvation and the hydrogen–air interface being under frequent start-stop conditions. Afterward, a quadratic function is built as the empirical degradation model according to the degradation evolution.

A model-based method is proposed to estimate the degradation degree α, degradation speed v and degradation acceleration a of PEMFCs stack by particle filter. Besides, a new health indicator of PEMFCs, rated voltage, is proposed, which can be used online not only under constant loading conditions but also under variable loading conditions. Based on this information, the degradation trend and RUL can be estimated online. Moreover, the real aging test data show the proposed prognostics method can predict the degradation trend and RUL at variable degradation accelerations, which has great application potential.

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Conflicts of Interest: The authors declare no conflict of interest.

Nomenclature

Abbreviations	
PEMFCs	Proton exchange membrane fuel cells
RUL	Remaining useful life
EOL	End-of-life
DOE	(USA) Department of Energy
EKF	Extended Kalman Filter
UKF	Unscented Kalman Filter
AUKF	Adaptive Unscented Kalman Filter
PF	Particle Filter
ECSA	Electrochemical surface area
MEA	Membrane electrode assembly
RMSE	Root mean square error
Physis symbols	
t_{pred}	The time to start the prediction (h)
t_{EOL}	The end-of-life time (h)
U_{avg}	The average voltage of the PEMFCs stack (V)
U_{stack}	Voltage of the PEMFCs stack (V)
E_0	Open circuit voltage (V)

$\mathcal{R}$	Gas constant (J mol^{-1} K^{-1})
T	Thermodynamic temperature (K)
F	Faraday constant (C mol^{-1})
j	Current density (A cm^{-2})
j_0	Exchange current density (A cm^{-2})
R	Ohmic resistance (Ω)
j_L	Limiting diffusion current density (A cm^{-2})
$j_0(t)$	The exchange current density at time t (A cm^{-2})
$R(t)$	The Ohmic resistance at time t (Ω)
α	Degradation degree
v	Degradation speed
a	Degradation acceleration
X_k	The health status of the PEMFCs stack at time k
Z_k	The average voltage of the PEMFCs stack at time k
ω	Process noise
φ	Observation noise
j_rated	The rated current density (A cm^{-2})
U_{α_k,j_k}	The real measured voltage at time k (V)
$U_{\alpha_k,j_\mathrm{rated}}$	The rated voltage at time k (V)

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