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Remaining Useful Life Prediction of Lithium-Ion Batteries Based on Wiener Processes with Considering the Relaxation Effect

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Abstract: Remaining useful life (RUL) prediction has great importance in prognostics and health management (PHM). Relaxation effect refers to the capacity regeneration phenomenon of lithium-ion batteries during a long rest time, which can lead to a regenerated useful time (RUT). This paper mainly studies the influence of the relaxation effect on the degradation law of lithium-ion batteries, and proposes a novel RUL prediction method based on Wiener processes. This method can simplify the modeling complexity by using the RUT to model the recovery process. First, the life cycle of a lithium-ion battery is divided into the degradation processes that eliminate the relaxation effect and the recovery processes caused by relaxation effect. Next, the degradation model, after eliminating the relaxation effect, is established based on linear Wiener processes, and the model for RUT is established by using normal distribution. Then, the prior parameters estimation method based on maximum likelihood estimation and online updating method under the Bayesian framework are proposed. Finally, the experiments are carried out according to the degradation data of lithium-ion batteries published by NASA. The results show that the method proposed in this paper can effectively improve the accuracy of RUL prediction and has a strong engineering application value.

Keywords: lithium-ion battery; relaxation; remaining useful life; regenerated useful time; Wiener processes; Bayesian framework; maximum likelihood estimation

1. Introduction

1.1. Motivations and Technical Challenges

Due to the long life, high energy density, fast charging speed, and high voltage, lithium-ion batteries have become an important research and development direction for energy storage batteries in a new energy field [1–3]. However, the safety and reliability for equipment highly depends on the health performance of energy storage batteries. As the main energy storage equipment, lithium-ion batteries will age gradually in actual use [4], which could lead to performance degeneration or even failure [5]. Failure of the lithium-ion batteries could result in inconvenience, equipment downtime, or even catastrophic accident [6]. Therefore, it is necessary to take measures to ensure the reliability and safety for lithium-ion batteries. Engineering practice shows that the prognostics and health management (PHM) can improve the equipment reliability by implementing necessary management actions before failure [7]. In PHM, remaining useful life (RUL) prediction can estimate the failure time and mitigate the risk for lithium-ion batteries by assessing the battery health [8], which is a key issue in order to make appropriate maintenance strategies and reduce accident risk [9]. However, due to the time-varying external environment and the complexity of internal electrochemical performance in practical application, the lithium-ion batteries' health degrades irregularly. This increases the difficulty for RUL prediction and leads to no universally accepted RUL prediction method [10]. From above reviews, the RUL prediction for lithium-ion batteries has become a critical issue in PHM of lithium-ion batteries [8].

The capacity generated by the full charge–discharge operational cycle is generally used as an appropriate feature to characterize the long-term degradation phenomenon for lithium-ion batteries [11]. Hence, in the existing literature, the capacity is usually used as the health indicator for lithium-ion batteries. The end of life for lithium-ion batteries is usually defined as the time when the battery capacity has faded beyond acceptable limits (typically 20%~30% of the rated capacity) [12]. Then, predicting the RUL for lithium-ion batteries could be transformed into estimating the time when its capacity crosses a predefined failure threshold. Therefore, the issue of RUL prediction can be converted to capacity estimation.

The relaxation effect is a typical feature during the degradation of lithium-ion batteries, which can be described as: When a lithium-ion battery rests for some time long enough during the degradation, it will lead to a recovery process for battery capacity, which increases the available capacity for next cycle. In actual use, the relaxation effect caused by long time rest has a great impact on the degradation law of lithium-ion batteries [13]. Recently, the relaxation effect is regarded as an important research direction for lithium-ion batteries in a review about RUL prediction based on a data-driven method [13]. To predict the RUL accurately, the relaxation effect should be considered in the actual degradation process for lithium-ion batteries. At present, the RUL prediction method with considering the relaxation effect has not been studied thoroughly. Therefore, to improve the safety and reliability for lithium-ion batteries, the RUL prediction method with considering the relaxation effect needs to be further studied.

1.2. Literature Review

The existing RUL prediction methods for lithium-ion batteries mainly include the adaptive filtering method, the artificial intelligence method, and the stochastic process modeling method. The adaptive filtering mainly includes Kalman filtering [14], extended Kalman filtering [15], unscented Kalman filtering [16,17], particle filtering [18–23], and unscented particle filtering [24,25], etc. Although the adaptive filtering has higher accuracy for RUL prediction, the accuracy can be easily influenced by time-varying current and ambient temperature [3]. The artificial intelligence method uses machine learning to fit the degradation path based on the monitored degradation data for RUL prediction. The typical artificial intelligence method includes support vector machine [26], support vector regression [27–29], relevance vector machine [30–32], relevance vector regression [17], neural network [33–35], genetic algorithm [36], particle swarm optimization algorithm [37], artificial fish swarm algorithm [38,39], and deep learning [40], etc. The artificial intelligent method can predict the non-linear system by using a simple algorithm and has a better prediction accuracy. However, these methods could only obtain the expectation for RUL, which cannot describe the uncertainty for the prediction results [3]. The stochastic process modeling method is based on probability theory, stochastic processes, and then the RUL can be predicted by establishing a stochastic degradation model. Since the stochastic process can well describe the uncertainty of the degradation process [41,42], many scholars have used the stochastic process to model the degradation of lithium-ion batteries for RUL prediction [41,43]. The degradation model based on the stochastic process mainly includes the Gaussian process and the Wiener process [3,44]. These methods can obtain the probability distribution function (PDF) of RUL, which could well describe the uncertainty of the degradation process for lithium-ion batteries.

The Wiener process is a type of diffusion process driven by Brownian motion with a drift coefficient. Since the Wiener process is suitable for describing the non-monotonic characteristic degradation processes with discontinuous increase or decrease trends, it has been widely applied in general degradation modeling [41]. Zhai et al. [45] predicted the RUL by using an adaptive Wiener process to model the degradation process for lithium-ion batteries. Tang et al. [43] developed a degradation model based on linear Wiener processes with measurement error for RUL prediction, which can well describe the capacity degradation process. Considering the nonlinear characteristic of the degradation process during the actual use for lithium-ion batteries, Si et al. [46] predicted the RUL by establishing a nonlinear Wiener process to model the capacity degradation. Wang et al. [47] predicted the RUL by developing a two-stage Wiener process method to model the degradation process for lithium-ion batteries. Feng et al. [48] proposed a RUL prediction method for lithium-ion batteries based on a two-dimensional Wiener process. It can be observed from above reviews that the Wiener process can better model the degradation process for lithium-ion batteries in actual use. Therefore, this paper mainly studies the RUL prediction method based on Wiener processes.

Recently, the influence of the relaxation effect on the degradation law of lithium-ion batteries has been considered in PHM. For modeling the relaxation effect, Saha and Goebel [49] modeled the relaxation effect based on an exponential model. Jin et al. [11] and Tang et al. [43] used this exponential function to extract the relaxation effect, and used the transformed data to predict the RUL. However, the relaxation effect is not considered in RUL prediction. Pei et al. [50] studied the relationship between rest time and the regenerated capacity in the degradation process. For RUL prediction with considering the relaxation effect, Qin et al. [51,52] proposed a prognostic framework with considering the relaxation effect for lithium-ion batteries, and used the beginning time interval of two adjacent cycles to reflect the rest time. Zhang et al. [53] proposed a RUL prediction method with recovery in storage, which has a higher accuracy for the lithium-ion batteries with state recovery. Zhang et al. [54] proposed a RUL prediction method with consideration of the relaxation effect, which uses the random jumps to describe the regeneration phenomena caused by the rest time. This method can better describe the recovery process caused by the relaxation effect. However, the calculation of RUL prediction is relatively complex. From the above reviews, it can be observed that the RUL prediction method with considering the relaxation effect has not been studied thoroughly. Therefore, to improve the safety and reliability for lithium-ion batteries in actual use, we study the RUL prediction method with consideration of the relaxation effect in this paper. What clearly distinguishes this work from the aforementioned literature is that we use the RUT to model the relaxation effect, which can lower the modeling complexity and reduce the difficulty for RUL prediction.

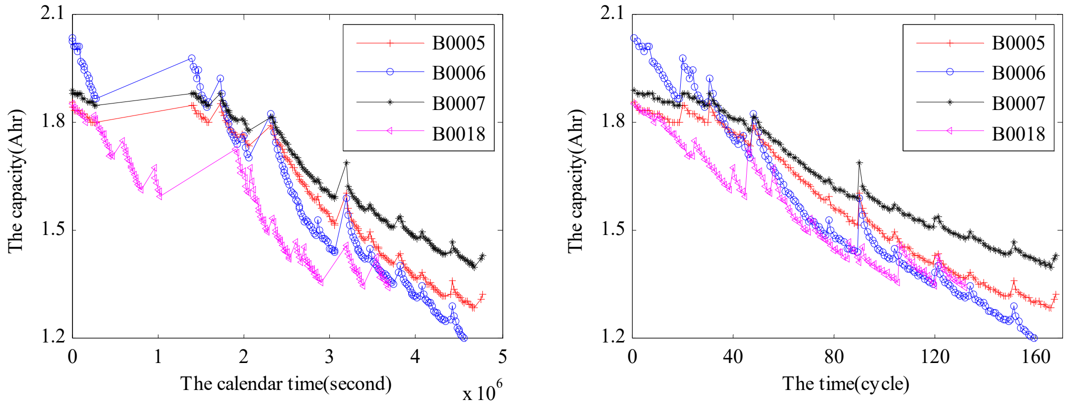
1.3. Original Contributions and Outline of Paper

There are two original contributions that clearly distinguish our endeavor from the aforementioned literature: The first important contribution of this paper is that the RUT caused by the relaxation effect is used to model the recovery process for lithium-ion batteries. This operation can simplify the modeling complexity and reduce the difficulty for RUL prediction with the relaxation effect. Another major contribution of this paper is that a novel RUL prediction method based on RUT and Wiener processes is proposed. This method can obtain the PDF of RUL and well describe the uncertainty of degradation process for lithium-ion batteries with relaxation effect.

The remainders of the paper are presented as follows: Section 2 analyzes the influence of the relaxation effect on the degradation law of lithium-ion batteries based on the degradation data presented by National Aeronautics and Space Administration (NASA). In Section 3, the degradation model that eliminates the relaxation effect is established based on Wiener processes, and an unbiased two-step maximum likelihood estimation (MLE) parameter estimation method is proposed. Section 4 develops a RUT model based on normal distribution, and proposes an MLE method for parameter estimation. In Section 5, a global RUL prediction method with considering the relaxation effect for lithium-ion batteries is developed by fusing the above two models. In Section 6, a practical case study of lithium-ion batteries is provided to illustrate the usefulness of the presented method.

2. Relaxation Effect Analysis

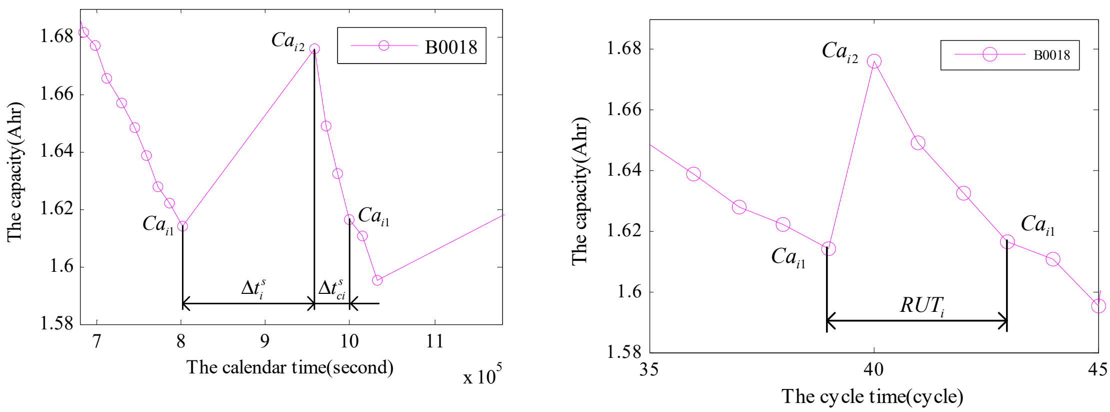
In this section, the relaxation effect is analyzed based on the degradation data of lithium-ion batteries published by NASA, as shown in Figure 1. The relaxation effect is a battery capacity regeneration phenomenon that occurs after a long period of rest. In Figure 1, the degradation data include two time scales, namely calendar time and number of cycles. For convenience, in this paper, t^s is used to indicate the calendar time, and t is the number of cycles. Each cycle t can be mapped to a specific calendar time t^s. The names (i.e., B0005, B0006, B0007, and B0018) in the legends denote the name of one specific battery. For more details, see [11,43].



(**a**) (**b**)

Figure 1. The experimental degradation data: (**a**) With calendar time; and (**b**) with cycle time.

Figure 1 shows that the battery capacity regeneration phenomenon occurs during some long rest time (for example, the calendar time interval (8×10^5, 10×10^5) of No. 18 battery), then the specific recovery process caused by relaxation effect can be expressed in Figure 2. Where Δt_1^s is the rest time, Ca_{i1} represents the beginning capacity of the relaxation effect, Ca_{i2} represents the capacity after a long rest time, $\mathrm{RUT_i}$ is the regenerated useful time caused by relaxation effect.



(**a**) (**b**)

Figure 2. The example of recovery process: (**a**) With calendar time; and (**b**) with cycle time.

From Figure 2a, a complete recovery process can be divided into two stages, The first stage is the capacity's regeneration process, where the capacity increases from Ca_{i1} to Ca_{i2} when the lithium-ion battery have a rest calendar time Δt_i^s. The second stage is the degradation process of the regenerated capacity. In this stage, the capacity drops from Ca_{i2} to Ca_{i1} after the end of break. The RUT caused by relaxation effect is shown in Figure 2b. From above works, a complete recovery process can be expressed as $U(ca_{i1} \to ca_{i2} \to ca_{i1} | \Delta t_i^s + t_{ci}^s)$ or $U(ca_{i1} \to ca_{i1} | RUT_i)$.

If all the rest times $\{\Delta t_1^s, \Delta t_2^s, \ldots \Delta t_m^s\}$ are known, all the recovery processes can be expressed as $\sum_{i=1}^{m} U(ca_{i1} \rightarrow ca_{i2} \rightarrow ca_{i1}|\Delta t_i^s + t_{ci}^s, RUT_i)$. The RUL prediction is different from the State-of-Health estimation, since it pays more attention to the remaining useful time. Therefore, the degradation model of recovery process (i.e., $\sum_{i=1}^{m} U(ca_{i1} \rightarrow ca_{i2} \rightarrow ca_{i1}|\Delta t_i^s + t_{ci}^s)$) is not needed to model, and we use the relationship between the RUT and the rest calendar time Δt_i^s to model the relaxation effect. As the capacity increasing process from Ca_{i1} to Ca_{i2} and the capacity degradation process of the regenerated capacity are not considered, this modeling method could lower the modeling complexity and reduce the difficulty for RUL prediction.

From the overall degradation trend of lithium-ion batteries, the relaxation effect could lead to an irregular degradation for battery capacity throughout the life cycle. If the relaxation effect is not considered in the degradation model, the accuracy of RUL prediction will be significantly reduced or even result in premature failure. Therefore, this paper mainly studies the influence of the relaxation effect on the degradation law of lithium-ion batteries, and proposes a novel online RUL prediction method with considering the relaxation effect. This method divides the degradation process of a lithium-ion battery life cycle into two parts. The first part is the degradation process that eliminates the recovery process, as shown in Figure 3. The nonlinear characteristic of this part is significantly reduced. The second part is the RUT caused by the relaxation effect, as show in Figure 2b.

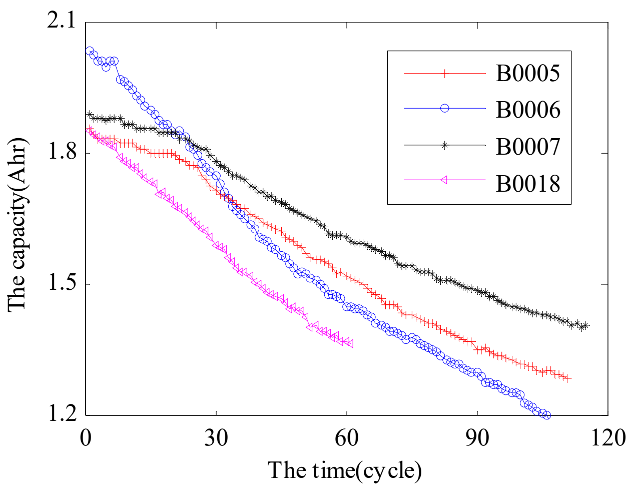


Figure 3. The degradation data after eliminating the relaxation effect.

3. RUL Prediction for the Degradation Model with Elimination of the Relaxation Effect

A method for the elimination of the relaxation effect and a RUL prediction method after eliminating the relaxation effect are proposed, respectively, in this section. The Wiener process is a type of diffusion process driven by Brownian motion with a drift coefficient, which is suitable for describing the non-monotonic degradation process with discontinuous increase or decrease trends. Furthermore, it is widely used in general degradation modeling [41]. It can be seen from Figure 3 that the linear characteristics of degradation for lithium-ion batteries are obvious after eliminating the relaxation effect. Therefore, the RUL prediction method proposed in this section is based on linear Wiener processes.

3.1. The Method for Eliminating the Relaxation Effect

We use a simple algorithm to eliminate the relaxation effect. The basic idea is to extract the relationship between the rest time and the regenerated capacity in calendar time scale, and extract the degradation process of the regenerated capacity until it degrades to the capacity value before regeneration. Then, it is mapped to the whole recovery process. The specific algorithm flow chart is shown in Figure 4.

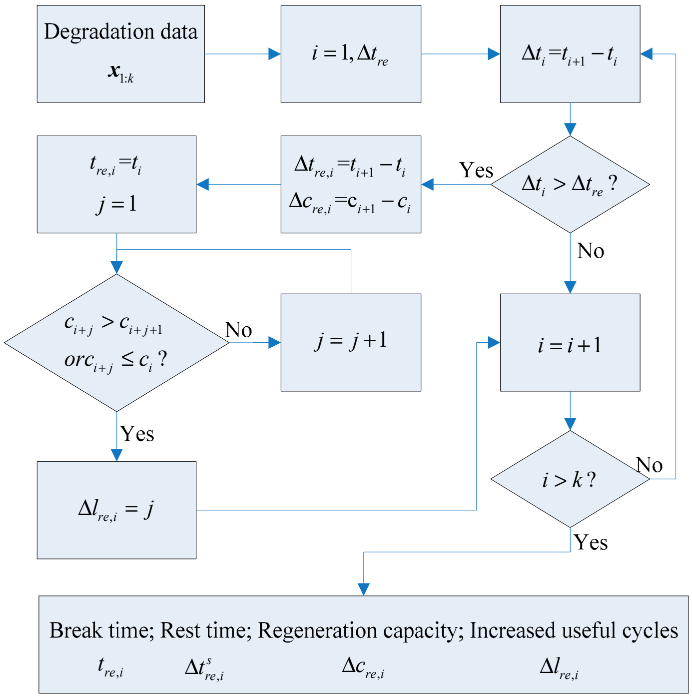


Figure 4. The algorithm flow chart for eliminating the recovery process.

In Figure 4, Δt^s_{re} is the minimum rest time with regeneration phenomenon for lithium-ion batteries. It is a subjective value given by analyzing the degradation process. According to this algorithm, all the information about the recovery process can be obtained; the rest time $t_{re,i}$, the rest time interval $\Delta t^s_{re,i}$, the regenerated capacity $\Delta c_{re,i}$ and the degradation cycle number $\Delta l_{re,i}$ of the regenerated capacity. Based on the above information, the relaxation effect of the lithium-ion batteries can be eliminated in the time interval $\sum [t_{re,i}, t_{re,i} + \Delta l_{re,i}]$. Then, the degradation process after eliminating the relaxation effect can be obtained, as shown in Figure 3.

3.2. Degradation Modeling

The degradation process of lithium-ion batteries after eliminating the relaxation effect can be expressed by the linear Wiener process as follows [43]:

$$X(t) = x_0 + \lambda t + \sigma_B B(t) \tag{1}$$

where λ is a drift coefficient that characterizes the degradation rate, σ_B is the diffusion coefficient, x_0 is the initial capacity, and $B(t)$ is the standard Brownian motion that represent the dynamic characteristics and uncertainty for the degradation process. In order to distinguish the individual difference among different batteries, drift coefficient λ is regarded as a random variable and follows normal distribution (i.e., $\lambda \sim N(\mu_\lambda, \sigma^2_\lambda)$).

The RUL based on the degradation process is defined as the time when the battery capacity cross the failure threshold at the first time. If the failure threshold of lithium-ion batteries is h, the RUL T of lithium-ion batteries can be defined as the time when the capacity reaches the failure threshold for the first time.

$$T = \inf\{t : X(t) \geq h | x_0 < h\} \tag{2}$$

3.3. Prior Parameters Estimation

In this subsection, an unbiased two-step MLE method is adopted to estimate the prior parameters. It is supposed that there are N batteries based on the historical degradation data, the detection time is $t_{n,1}, t_{n,2}, \ldots t_{n,m_n}$, where m_n represents the number of detections of the No. n battery, the true degradation data of the detection time $t_{n,i}$ is $x_{n,i} = X(t_{n,i})$, the degradation data of all batteries are $\{\boldsymbol{x}_{1:m_1}, \boldsymbol{x}_{1:m_2}, \ldots, \boldsymbol{x}_{1:m_N}\}$, where $\boldsymbol{x}_{1:m_n} = \{x_{n,1}, x_{n,2}, \ldots, x_{n,m_n}\}$. Let $\Delta x_{n,i} = x_{n,i} - x_{n,i-1}$, $\Delta t_{n,i} = t_{n,i} - t_{n,i-1}$, then according to the property of Wiener processes, $\Delta x_{n,i}$ obeys a normal distribution (i.e., $\Delta x_{n,i} \sim N(\lambda_n \Delta t_{n,i}, \sigma_{B1}^2 \Delta t_{n,i})$), where λ_n represents the drift coefficient of the No. n battery. As $\Delta t_{n,i} = 1$, we can obtain that $\Delta x_{n,i} \sim N(\lambda_n, \sigma_{B1}^2)$. For general linear Wiener processes, the unknown parameters are $\boldsymbol{\psi} = \{\mu_\lambda, \sigma_\lambda^2, \sigma_B^2\}$, Since the drift coefficient $\boldsymbol{\lambda} = \{\lambda_1, \lambda_2, \ldots, \lambda_N\}$ of each battery is unknown when estimating $\boldsymbol{\psi}$, this section estimates the unknown parameters by using a two-step method presented by Tang et al. [43]. The specific algorithm is given as follows:

Step 1: Establishing the log-likelihood function for the degradation data of N lithium-ion batteries with respect to σ_B^2 and $\boldsymbol{\lambda}$, and estimating σ_B^2 and $\boldsymbol{\lambda}$ by the MLE method.

First, the log-likelihood function can be written as follows:

$$\ln L(\boldsymbol{\lambda}, \sigma_B^2 | \sum_{n=1}^{N} \boldsymbol{x}_{1:m_n}) = \sum_{n=1}^{N} \left(-\frac{m_n(\ln 2\pi + \ln \sigma_B^2)}{2} - \frac{\sum_{i=1}^{m_n} (\Delta x_{n,i} - \lambda_n)^2}{2\sigma_B^2} \right) \tag{3}$$

Take the partial derivatives of λ_n and σ_B^2 for (3):

$$\frac{\partial}{\partial \lambda_n} \ln L(\lambda_n, \sigma_B^2 | \boldsymbol{x}_{1:m_n}) = -\frac{1}{\sigma_B^2} \sum_{i=1}^{m_n} (\lambda_n - \Delta x_{n,i}) \tag{4}$$

$$\frac{\partial}{\partial \sigma_B^2} \ln L(\lambda_n, \sigma_B^2 | \sum_{n=1}^{N} \boldsymbol{x}_{1:m_n}) = \sum_{n=1}^{N} \left(-\frac{m_n}{2\sigma_B^2} + \frac{1}{2\sigma_B^4} \sum_{i=1}^{m_n} (\Delta x_{i,n} - \lambda_n)^2 \right) \tag{5}$$

Then, the MLE for λ_n and σ_B^2 can be obtained by setting (4) and (5) to zero. That is:

$$\lambda_n = \frac{x_{n,m_n}}{m_n} \tag{6}$$

$$\sigma_B^2 = \frac{\sum_{n=1}^{N} \sum_{i=1}^{m_n} (\Delta x_{n,i} - \lambda_n)^2}{\sum_{n=1}^{N} m_n} \tag{7}$$

Step 2: Calculating the prior distribution of the drift coefficients based on the estimation of $\boldsymbol{\lambda} = \{\lambda_1, \lambda_2, \ldots, \lambda_N\}$ in the first step.

According to the result of $\boldsymbol{\lambda} = \{\lambda_1, \lambda_2, \ldots, \lambda_N\}$ in step 1, the prior distribution of $\boldsymbol{\lambda}$ can be obtained as follows:

$$\mu_\lambda = \frac{1}{N} \sum_{i=1}^{N} \lambda_i, \sigma_\lambda^2 = \frac{1}{N} \sum_{i=1}^{N} (\lambda_i - \mu_\lambda)^2 \tag{8}$$

In general, the number of lithium-ion batteries used in the degradation experiments is relatively small, the results of σ_λ^2 could be underestimated by (8). Therefore, a modified unbiased estimation method is proposed for the drift coefficient. That is:

$$\sigma_\lambda^2 = \frac{1}{N-1} \sum_{i=1}^{N} (\lambda_i - \mu_\lambda)^2 \tag{9}$$

3.4. Online Parameter Updating and RUL Prediction

Define that $\boldsymbol{x}_{1:k} = \{x_1, x_2, \ldots, x_k\}$ are the degradation data for a lithium-ion battery at time $t_1, t_2, \ldots t_k$, then given the prior distribution of the drift coefficient λ, the posterior distribution of λ conditional on $\boldsymbol{x}_{1:k}$ also follows the normal distribution according to Bayesian theory [55]. That is:

$$\lambda | \boldsymbol{x}_{1:k} \sim N(\mu_{\lambda,k}, \sigma_{\lambda,k}^2) \tag{10}$$

where
$$\mu_{\lambda,k} = \frac{x_k\sigma_\lambda^2 + \mu_\lambda\sigma_B^2}{t_k\sigma_\lambda^2 + \sigma_B^2}, \sigma_{\lambda 1,k}^2 = \frac{\sigma_B^2\sigma_\lambda^2}{t_k\sigma_\lambda^2 + \sigma_B^2} \tag{11}$$

After detecting the degradation data $\boldsymbol{x}_{1:k}$ of the lithium-ion battery, the degradation process when $t > t_k$ can be written as follow:

$$X(t|\boldsymbol{x}_{1:k}) = x_k + \mu_1(t - t_k) + \sigma_B B(t - t_k) \tag{12}$$

where $\mu_1 \sim N(\mu_{\lambda,k}, \sigma_{\lambda,k}^2)$.

Let $l_k = t - t_k(l_k \geq 0)$, then (12) can be transformed as follow:

$$Y(l_k) = X(l_k + t_k) - X(t_k) = \lambda l_k + \sigma_B B(l_k) \tag{13}$$

where $Y(0) = 0$.

According to (2), the RUL at time t_k can be transformed into the time when the degradation process $\{Y(l_k), l_k \geq 0\}$ first cross the failure threshold $w_k = w - x_k$. Then the corresponding RUL can be defined as follows:

$$L_k = \inf\{l_k : X(l_k + t_k) \geq w|\boldsymbol{x}_{1:k}\} = \inf\{l_k : Y(l_k) \geq w - x_k|\boldsymbol{x}_{1:k}\} \tag{14}$$

Therefore, the PDF of the RUL for lithium-ion batteries after eliminating the relaxation effect can be written as follows [55]:

$$f_{L_k|\boldsymbol{x}_{1:k},w}(l_k|\boldsymbol{x}_{1:k}, w) = \frac{w - x_k}{\sqrt{2\pi l_k^2(\sigma_{\lambda,k}^2 l_k^2 + \sigma_B^2 l_k)}} \exp\left(-\frac{(w - x_k - \mu_{\lambda,k}l_k)^2}{2(\sigma_{\lambda,k}^2 l_k^2 + \sigma_B^2 l_k)}\right) \tag{15}$$

4. RUT Prediction for the Relaxation Effect

This section focuses on the prediction of RUT caused by relaxation effect of lithium-ion batteries. The data of the relaxation effect is extracted by using the elimination algorithm proposed in Section 3.1.

4.1. Modeling the RUT

Define the regenerated useful time (RUT) as the increased RUL caused by the relaxation effect of lithium-ion batteries. By extracting the relaxation effect of the same type of lithium-ion batteries, the data between the rest time and the RUT can be obtained as shown in Figure 5.

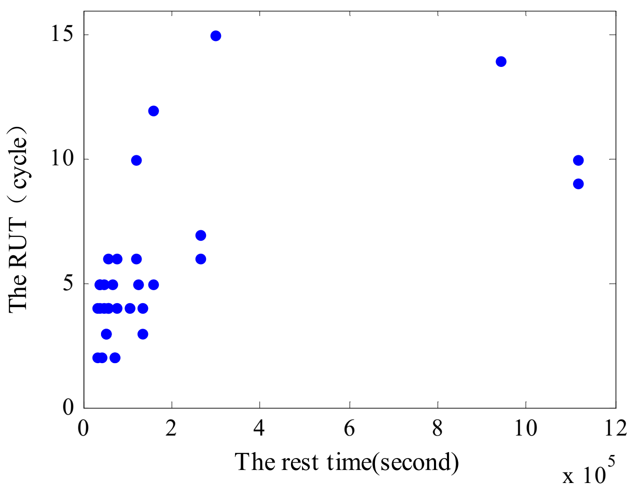


Figure 5. Data between the rest time and RUT.

From Figure 5, we observe that there is a hidden nonlinear relationship between the rest time and RUT. In order to better describe the uncertainty of this relationship, we model the RUT based on a normal distribution. It is assumed that the mean of RUT after the rest time Δt_i^s is $g(\Delta t_i^s)$. Here, $g(\Delta t^s) = a(\Delta t^s)^b$ is used to indicate the nonlinear relationship between the rest time and the RUT. Then, the RUT after a given rest time $\Delta t_{re,i}^s (i = 1, 2, \ldots, n)$ can be written as follows:

$$RUT_i \sim N(g(\Delta t_{re,i}^s), \sigma_r^2) \tag{16}$$

where $N(\cdot)$ represents the normal distribution, and σ_r^2 represents the uncertainty of the RUT caused by the rest time. Therefore, the unknown parameters for the RUT model are $\boldsymbol{\Theta} = \{\sigma_r^2, a, b\}$.

4.2. Parameters Estimation

In this subsection, the unknown parameters are estimated by using the MLE method. The log-likelihood function based on the data information of rest time $\Delta \boldsymbol{t}_{re,1:n}^s = \{\Delta t_{re,1}^s, \Delta t_{re,2}^s, \ldots, \Delta t_{re,n}^s\}$ and RUT $\boldsymbol{r}_{1:n} = \{r_1, r_2, \ldots, r_n\}$ can be written as follows:

$$\ln L(\boldsymbol{\Theta}|\boldsymbol{r}_{1:n}) = -\frac{n}{2}(\ln 2\pi + \ln \sigma_r^2) - \frac{1}{2\sigma_r^2}\sum_{i=1}^{n}(r_i - g(\Delta t_{re,i}^s))^2 \tag{17}$$

Taking the partial derivatives of a and σ_r^2 for (17) gives:

$$\frac{\partial}{\partial a}\ln L(\boldsymbol{\Theta}|\boldsymbol{r}_{1:n}) = \frac{1}{\sigma_r^2}\sum_{i=1}^{n}\Big((\Delta t_{re,i}^s)^b\Big(a(\Delta t_{re,i}^s)^b - r_i\Big)\Big) \tag{18}$$

$$\frac{\partial}{\partial \sigma_r^2}\ln L(\boldsymbol{\Theta}|\boldsymbol{r}_{1:n}) = -\frac{n}{2\sigma_r^2} + \frac{1}{2\sigma_r^4}\sum_{i=1}^{n}(r_i - g(\Delta t_{re,i}^s))^2 \tag{19}$$

By setting (18) and (19) equal to 0, the restricted estimation for a and σ_r^2 limited by b can be obtained as follows:

$$\hat{a} = \frac{\sum_{i=1}^{n}\Big((\Delta t_{re,i}^s)^b r_i\Big)}{\sum_{i=1}^{n}(\Delta t_{re,i}^s)^{2b}} \tag{20}$$

$$\hat{\sigma}_r^2 = \frac{1}{n}\sum_{i=1}^{n}\left(r_i - \frac{\sum_{i=1}^{n}\Big((\Delta t_{re,i}^s)^b r_i\Big)}{\sum_{i=1}^{n}(\Delta t_{re,i}^s)^{2b}}(\Delta t_{re,i}^s)^b\right)^2 \tag{21}$$

Substituting (20) and (21) into log-likelihood function (17), and simplifying, gives the profile log-likelihood function for b in terms of the estimated (a, σ_r^2) as:

$$\ln L(b|\boldsymbol{r}_{1:n}) = -\frac{n}{2}\ln 2\pi - \frac{n}{2}\ln\left[\frac{1}{n}\sum_{i=1}^{n}\left(r_i - \frac{\sum_{i=1}^{n}\Big((\Delta t_{re,i}^s)^b r_i\Big)}{\sum_{i=1}^{n}(\Delta t_{re,i}^s)^{2b}}(\Delta t_{re,i}^s)^b\right)^2\right] - \frac{n}{2} \tag{22}$$

The estimation of b can be obtained by maximizing (22).

$$\hat{b} = \underset{b}{\operatorname{argmax}} \ln L(b|\boldsymbol{r}_{1:n}) \tag{23}$$

Then the estimation of a and σ_r^2 can be obtained by substituting b into (20) and (21). The "Fminsearch" function in MATLAB is used to search for the estimated value of b.

4.3. Predicting the RUT

From the above works, the issue of RUL prediction for the relaxation effect can be transformed into the RUT prediction with the rest time $\Delta t^s_{re,i}(i = 1, 2, \ldots, n)$. When all the rest time of a lithium-ion battery is known to be $\Delta t^s_{re,i}(i = 1, 2, \ldots, n)$ during its life cycle, the RUT caused by the rest time can be written as follows:

$$RUT \sim N\left(\sum_{i=1}^{n} g(\Delta t^s_{re,i}), n\sigma_r^2\right) \tag{24}$$

In conclusion, at the current time k, the RUT caused by the rest time in the future can be expressed as follows:

Predicting the RUT based on conditional probability if the current time k is in the recovery process [43]. Let l_m denote the cycle time already used in the current recovery process $U(ca_{m1} \to ca_{m1}|RUT_m)$, $\phi(\cdot)$ denote the probability density function of the standard normal distribution, $\Phi(\cdot)$ denote the cumulative distribution function of the standard normal distribution. Then the PDF of RUT_1 in the current recovery process can be written as follows:

$$f(RUT_1) = \frac{1}{\Phi((g(\Delta t_{re,m}) - l_m)/\sigma_r)\sqrt{2\pi\sigma_r^2}} \cdot \exp\left[-\frac{(RUT_1 - (g(\Delta t_{re,m}) - l_m))^2}{2\sigma_r^2}\right] \tag{25}$$

where:

$$E(RUT_1) = g(\Delta t_{re,m}) - l_m - \frac{\sigma_r}{\sqrt{2\pi}\Phi((g(\Delta t_{re,m}) - l_m)/\sigma_r)} \cdot \exp\left[-\frac{(g(\Delta t_{re,m}) - l_m)^2}{2\sigma_r^2}\right] \tag{26}$$

$$\mathrm{var}(RUT_1) = \sigma_r^2\left[1 - \frac{(g(\Delta t_{re,m}) - l_m)/\sigma_r)\phi((g(\Delta t_{re,m}) - l_m)/\sigma_r)}{\Phi((g(\Delta t_{re,m}) - l_m)/\sigma_r)} - \left(\frac{\phi((g(\Delta t_{re,m}) - l_m)/\sigma_r)}{\Phi((g(\Delta t_{re,m}) - l_m)/\sigma_r)}\right)^2\right] \tag{27}$$

Else if the current time k is not in the recovery process, then the PDF of RUT_1 can be written as follows:

$$f(RUT_1) = 0 \tag{28}$$

where:

$$E(RUT_1) = 0, \quad \mathrm{var}(RUT_1) = 0 \tag{29}$$

When all the rest time of a lithium-ion battery are known to be $\Delta t^s_{re,i}(i = m + 1, m + 2, \ldots, n)$ during its late life cycle, the PDF of RUT_2 caused by the rest time can be written as follows:

$$f(RUT_2) = \frac{1}{\sqrt{2\pi(n - m)\sigma_r^2}} \exp\left[-\frac{\left(RUT_2 - \sum_{i=m+1}^{n} g(\Delta t^s_{re,i})\right)^2}{2(n - m)\sigma_r^2}\right] \tag{30}$$

where:

$$E(RUT_2) = \sum_{i=m+1}^{n} g(\Delta t^s_{re,i}), \mathrm{var}(RUT_2) = (n - m)\sigma_r^2 \tag{31}$$

From the above reviews, the RUT caused by the relaxation effect can be expressed as the convolution of RUT_1 and RUT_2. Then the PDF of RUT can be written as follows:

$$f(RUT) = f(RUT_1) * f(RUT_2) \tag{32}$$

5. The Global RUL Prediction for Lithium-Ion Batteries with Considering the Relaxation Effect

In this section, a novel global RUL prediction method with considering the relaxation effect is developed. For simplicity, suppose that R_1 represents the RUL that eliminates the relaxation effect,

R_2 represents the RUT that is caused by the relaxation effect, and R is the global RUL of lithium-ion batteries. From above works, the PDF of R_1 that eliminates the relaxation effect can be obtained as follows:

$$f(R_1) = \frac{w - x_k}{\sqrt{2\pi {R_1}^2(\sigma_B^2 R_1 + \sigma_{\lambda,k}^2 {R_1}^2)}} \cdot \exp\left(-\frac{(w - x_k - \mu_{\lambda,k}R_1)^2}{2(\sigma_B^2 R_1 + \sigma_{\lambda,k}^2 {R_1}^2)}\right) \tag{33}$$

Then, the PDF of R_2 that is caused by the relaxation effect can be obtained as follows:

$$f(R_2) = f(RUT_1) * f(RUT_2) \tag{34}$$

The basic idea of this method is to obtain the PDF of the R_1 that eliminates the relaxation effect and the PDF of R_2 that is caused by relaxation effect according to the degradation stage of lithium-ion batteries at the current time first. Then the PDF of the RUL for lithium-ion batteries can be obtained by convoluting R_1 and R_2. And this method is mainly divided into four steps. The specific algorithm flow is shown in Figure 6.

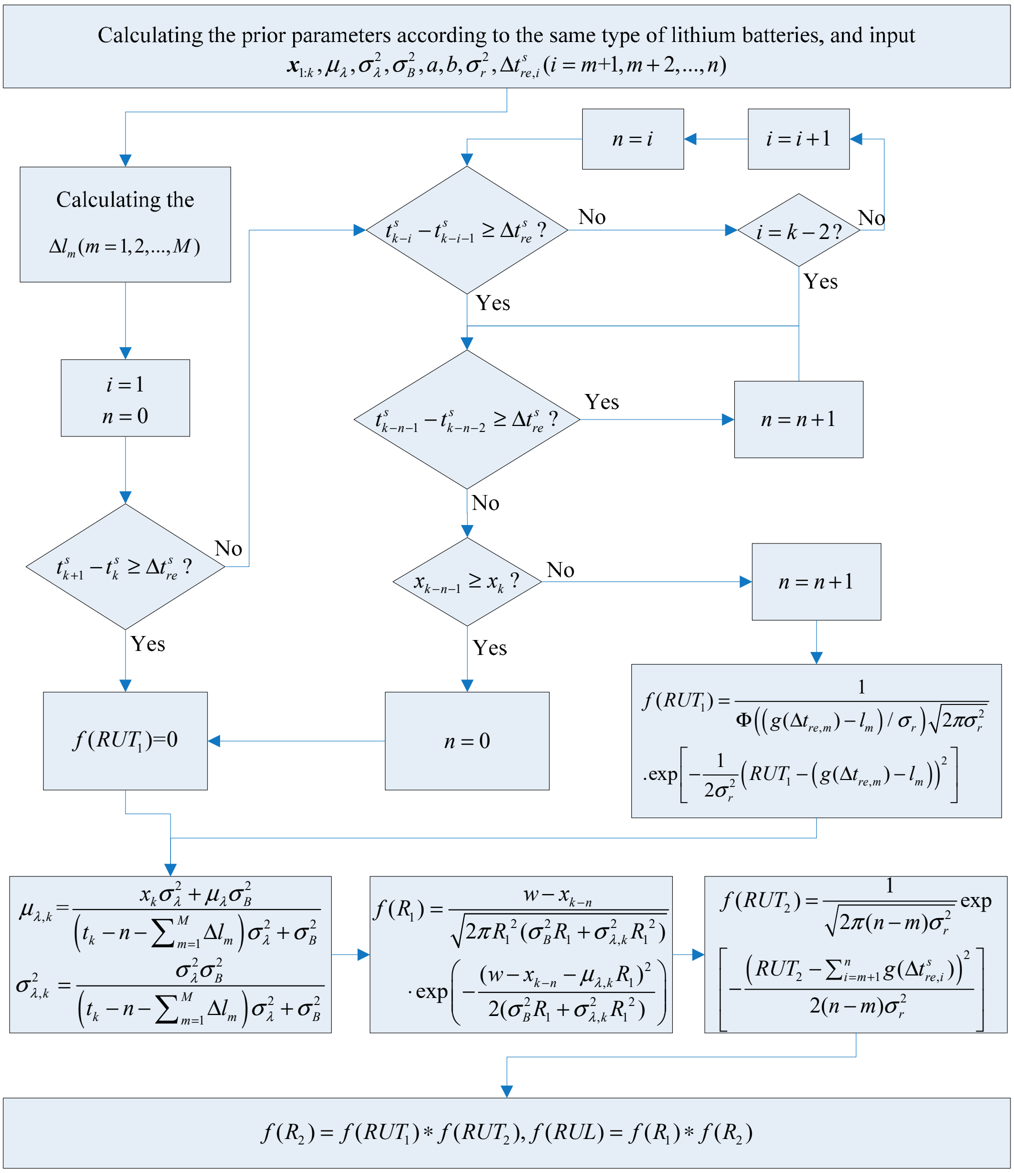


Figure 6. The algorithm flow chart for RUL prediction with considering the relaxation effect.

Step 1: Establishing the degradation model that eliminates the relaxation effect based on Wiener processes and the RUT model caused by the relaxation effect based on normal distribution.

Step 2: For the model based on the Wiener process, estimating the prior parameters by using the unbiased two-step MLE method, and then online updating the random coefficient under the Bayesian framework. For the model of RUT, estimating the parameters based on the MLE method.

Step 3: According to the on-site degradation data at the current time k, calculating the R_1 and R_2.

Step 4: The PDF of RUL considering the relaxation effect for lithium-ion batteries can be obtained by convoluting R_1 and R_2. That is

$$f(R) = f(R_1) * f(R_2) \tag{35}$$

6. Experiment

In this experiment, a practical case study of lithium-ion batteries based on the degradation data published by NASA was carried out to verify the effectiveness of the proposed RUL prediction method. The failure of lithium-ion batteries was defined as the time when the full charge capacity was reduced to below 70% of its rated value. For simplicity, the method proposed in this paper is referred to as M1, the method without the modeling relaxation effect as M2. The B0005 battery was chosen to compare these two methods, and the degradation data of other batteries were used to estimate the prior parameters. In order to better illustrate the estimation accuracy, we defined the relative error (RE) as shown in (36), which is an absolute value for the predicted RUL minus real RUL; the mean square error (MSE) as shown in (37), which is the definite integral that the PDF of predicted RUL multiply the square for the predicted RUL minus real RUL; the mean absolute percentage error (MAPE) as shown in (38), which is the average value for the RE from the current time t_k to predicted failure time; and the root mean square error (RMSE) as shown in (39), which is the square root value for the average of the MSE from the current time t_k to predicted failure time.

$$RE = |RUL_{real} - RUL_{es}| \tag{36}$$

$$MSE = \int_0^\infty (RUL_{real} - RUL_{es})^2 f_{RUL_{es}|\boldsymbol{x}_{1:k}}(RUL_{es}|\boldsymbol{x}_{1:k})dRUL_{es} \tag{37}$$

$$MAPE = \frac{1}{N}\sum_{i=k}^{N} |RUL_{i,real} - RUL_{i,es}| \tag{38}$$

$$RMSE = \sqrt{\frac{1}{N}\sum_{i=k}^{N}\int_0^\infty (RUL_{k,real} - RUL_{k,es})^2 f_{RUL_{k,es}|\boldsymbol{x}_{1:k}}(RUL_{k,es}|\boldsymbol{x}_{1:k})dRUL_{k,es}} \tag{39}$$

6.1. Prior Parameters Estimation for the Data with Elimination of the Relaxation Effect

The degradation data with Elimination of the Relaxation Effect of B0005, B0006, B0007, and B0018 lithium-ion batteries are as shown in Figure 3. It can be observed that the linear trend of the transformed data is more obvious than the original data. According to the transformed degradation data (without considering the B0005 battery), the prior parameter $\boldsymbol{\Psi}$ can be obtained based on the unbiased two-step MLE method presented in Section 3, as shown in Table 1.

Table 1. The estimation result of the prior parameter $\boldsymbol{\Psi}$.

μ_λ	σ_λ^2	σ_{B1}^2
−0.0063	2.6877×10^{-6}	2.9348×10^{-5}

6.2. Parameters Estimation for the Model of RUT

It can be observed from Figure 1 that when the rest time satisfied $\Delta t_i \geq 3 \times 10^4$, the battery capacity increased significantly. Therefore, it is considered that the rest time 3×10^4 is sufficient to produce a relaxation effect. According to the data between the rest time and the RUT, the prior parameter $\mathbf{\Theta}$ can be obtained based on the MLE method presented in Section 4, as shown in Table 2.

Table 2. The estimation result of the prior parameter $\mathbf{\Theta}$.

σ_r^2	a	b
4.9055	0.0139	0.5184

According to the parameters estimation result, the relationship function $g(t)$ between the rest time and the RUT can be obtained as shown in Figure 7.

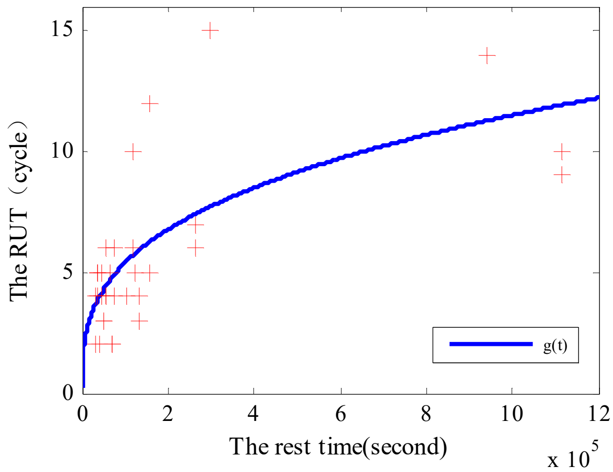


Figure 7. The relationship function $g(t)$ between the rest time and the RUT.

6.3. RUL Prediction

In the following, the RUL at all different time points are calculated, and we plot the estimated PDF of RUL and the actual RUL at some different time points in Figure 8. It shows from Figure 8 that the mode of the distribution by M2 is larger than that by M1, and the expectation of M1 is closer to the true RUL curve than M2. This indicates that the failure is more likely to happen earlier, which could cause premature maintenance and increase the maintenance costs. The reason for this phenomenon is that not extracting the relaxation effect could increase the degradation uncertainty, which results in overestimate of σ_B^2. Then, the mode of PDF is underestimated on the concept of first hitting time. Therefore, the relaxation effect needs to be modeled separately with the degradation and considered for RUL estimation.

Then, we calculate the results of RE, as shown in Figure 9a, and MSE, as shown in Figure 9b, at some time points. It can be observed that M1 obtains better accuracy than M2. From Figure 9a, it shows that M1 is closer to the actual RUL than M2, the RE of M1 is less than 2; however, the RE of M2 can be up to 25. From Figure 9b, it can be observed that not considering the relaxation effect could increase the degradation uncertainty, which results in overestimation of the σ_B^2.

Additionally, we calculate the MAPE, as shown in Figure 10a, and RMSE, as shown in Figure 10b, at some time points. It shows that M1 obtains better accuracy than M2. According to Figure 10a, the average value of the MAPE obtained by M1 is 1.01 and M2 is 3.14. This result indicates that the method proposed in this paper has high predicted accuracy. According to Figure 10b, the average value of RMSE obtained by the RUL prediction method can be obtained that M1 is 3.78 and M2 is 22.32.

This result shows that the proposed method by this paper not only has high accuracy but also has small uncertainty.

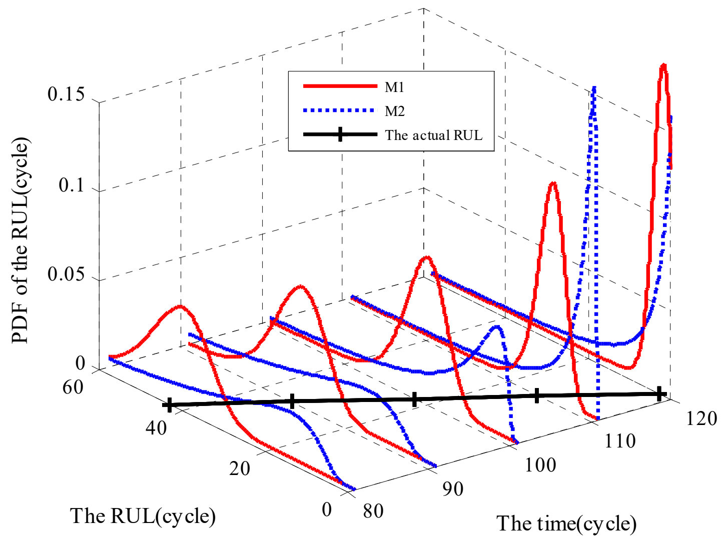


Figure 8. The estimated PDF of the RUL by M1 and M2.

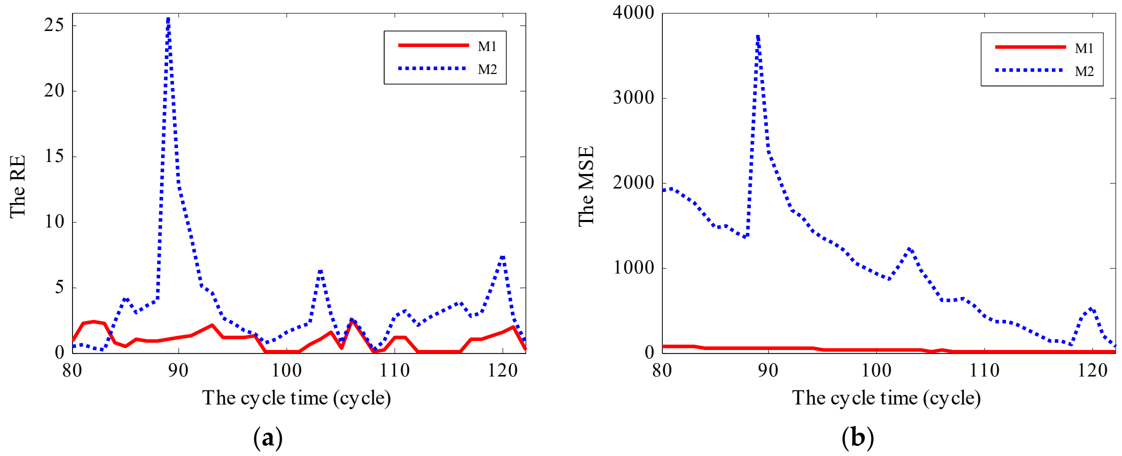


Figure 9. The results of RE and MSE at some time points: (**a**) The RE by M1 and M2; and (**b**) the MSE by M1 and M2.

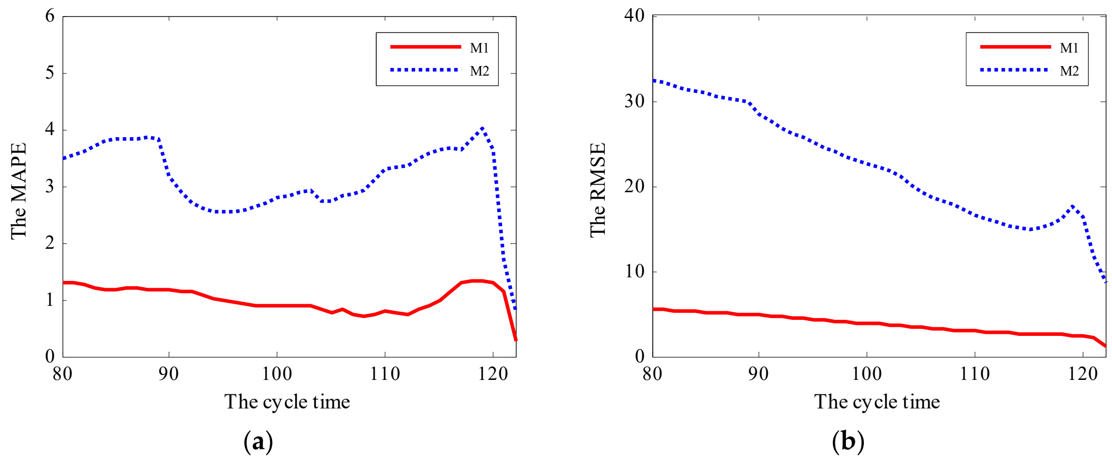


Figure 10. The results of MAPE and RMSE at some time points: (**a**) The MAPE by M1 and M2; and (**b**) the RMSE by M1 and M2.

7. Conclusions

The above works show that the RUL prediction with considering the relaxation effect is of great importance in PHM of lithium-ion batteries. In this paper, a novel online RUL prediction method is proposed by studying the relaxation effect on the degradation law of lithium-ion batteries, and the experiments are carried out based on the degradation data of lithium-ion batteries published by NASA. The results show the effectiveness and better accuracy of the proposed method by comparing with the method of that not considering the relaxation effect. Then the conclusions can be summarized as follows. Firstly, the relaxation effect has a big impact on the degradation law of lithium-ion batteries, thus it is necessary to model the relaxation effect to improve the accuracy of RUL prediction. The modeling difficult can be reduced by using the RUT to model the recovery process. Since the long rest time of lithium-ion batteries is not common in practical use, this method has strong engineering application value. Secondly, the RUL prediction method based on RUT and Wiener processes can obtain the PDF of RUL, which can well describe the uncertainty of lithium-ion batteries' degradation process.

A case study regarding practical data of lithium-ion batteries shows that the proposed model could effectively model the relaxation effect and has a better accuracy of RUL estimation than the existing method.

Author Contributions: X.X. completed the theoretical results and experiments, and wrote the paper; S.T. formulated the framework and completed the theoretical analysis; C.Y. structured the whole manuscript and revised the paper; X.S. (Xiaoyan Sun) formulated the framework and checked the experiments results. X.S. (Xiaosheng Si) checked the theoretical results and revised the paper; and L.W. revised the paper.

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