

Technology for Lower Limit Prediction of Lithium-ion Batteries Performance after Degradation Employing Diffusion Process Model

Kaoru OMICHI¹Kotone SUZUKI¹Hideki NAGATSUKA, Ph.D.²Michihiro YASUDA³

ABSTRACT

A model for predicting lower limit performance including variation terms was constructed to help achieve performance after durability testing of products equipped with lithium-ion batteries. To predict degradation, the model formula includes two types of stochastic variables: random effects multiplied by the battery durability degradation trend term, and a diffusion process in which variation expands proportionally with time. Parameter optimization was conducted by the maximum likelihood method aided by the Markov Chain Monte Carlo method. As a result, the lower limit performance after degradation, which had conventionally been predicted based on 12 months of measured durability data, was successfully computed based on only four months of durability data. It was also demonstrated that prediction of lower limit performance, which previously required the two pieces of information of the predicted typical performance values after durability testing and performance variation during durability, could be achieved with just one piece of information by using the proposed model.

1. Introduction

Lithium-ion batteries (LiB) have high energy density and power density, and their use is expanding from small electronic devices to large mobility applications⁽¹⁾⁻⁽⁵⁾. LiB for electric vehicles such as hybrid electric vehicles and battery electric vehicles need to provide durability over long periods of 10 years or more in consideration of diverse regions and applications⁽⁶⁾. To verify durability under various usages, it is more efficient to utilize simulation models rather than conducting individual tests using actual vehicles^{(7), (8)}. The authors previously constructed a model capable of efficiently predicting battery performance after degradation in a short time. In that study, it was found that, by training the model to learn the degradation behavior of the battery under varying load conditions and predicting degradation through accumulation of the degradation amount, nonlinear degradation behavior could be predicted^{(9), (10)}. However, this method was able to predict only the average performance value after degradation. On the other hand, the EU Battery Regulation sets minimum

performance requirements throughout the lifetime⁽⁶⁾, making it necessary to verify the LiB lower limit performance in the development phase. The lower limit performance can be predicted by subtracting the variation in Fig. 1(b) from the values predicted by the model in Fig. 1(a), as shown in Fig. 1(c). That is, it needs to be done in two steps: prediction by the degradation model and prediction of lower limit performance. Durability testing is needed in both steps, making this method both costly and time-consuming. This study aimed to address the aforementioned issue efficiently in a single step by employing the model illustrated in Fig. 1(d).

To achieve this goal, it is necessary to introduce terms into the degradation model formula that appropriately express the variation in performance after durability testing. This study employs a degradation model formula incorporating multiple random terms based on factors resulting in LiB performance variance after durability tests. This method has the issues that the number of parameters increases as random terms are introduced, which can lead

¹ Innovative Research Excellence, Power Unit & Energy

E-mail: kaoru_omichi@jp.honda

² Chuo University

³ TechnoPro, Inc. TechnoPro Design Company

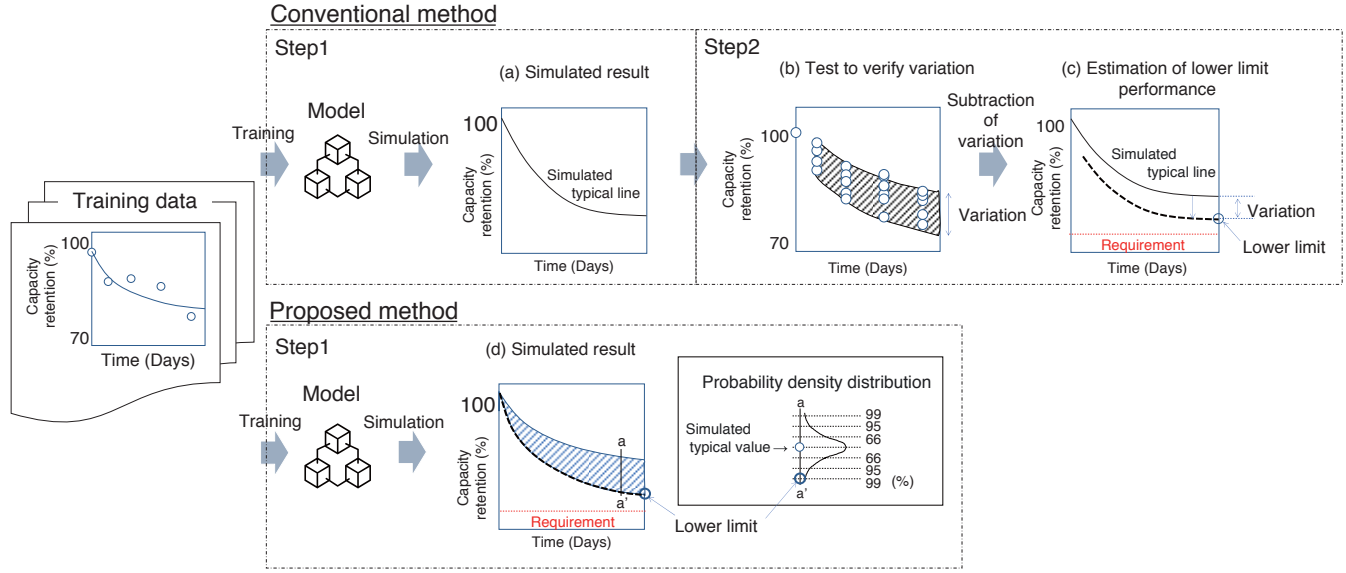


Fig. 1 Estimation of lower limit performance by conventional and proposed methods

to local optimization of parameters, and that the durability testing period needed for model training is unclear. Efforts were therefore made to address these issues.

2. Proposal of Degradation Model for Predicting Lower Limit

2.1. Consideration of Factors Resulting in Performance Variation

The remaining useful capacity after durability testing was analyzed to determine the random terms that represent variation. The battery type and durability test conditions were kept constant, and durability tests were conducted on multiple batteries to measure the changes in capacity over time. 5 Ah-class cylindrical batteries were used as test samples. Figure 2 shows the degradation behavior of 16 batteries charged and discharged from 4.1 V to 3.4 V at an ambient temperature of 55°C. The solid lines represent the behavior of capacity retention rate, and the circle plots indicate the variance of the capacity retention rate at each measurement point. As the variance tends to increase over time, there is individual variation in the degradation speed among batteries. Additionally, even for a single battery, the capacity retention rate fluctuates depending on the measurement timing, so there is measurement variation that is independent of the elapsed time. These two types of variation are present in LiB degradation behavior.

In order to construct a model based on varied performance, the parameters of the model will also include stochastic variation. As an example, Fig. 3 shows the observed degradation speed at 25, 40, and 55°C. The gray-shaded range shows the 95% confidence interval of a degradation model that has been trained on this degradation behavior. That is to say, the results output by a model

trained on varied data will also contain stochastic variation, which is referred to hereinafter as model error.

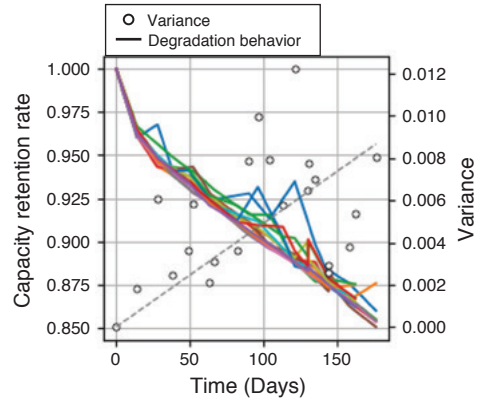


Fig. 2 Degradation behavior of LiB at same loading condition

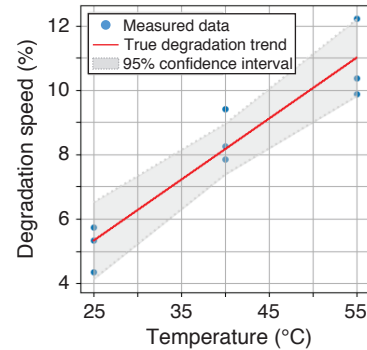


Fig. 3 Observed degradation speed and confidence interval

2.2. Formulation of Degradation Model for Predicting Lower Limit

The measurement variation of the measurement data in the LiB durability tests, the variation of durability performance, and the model error when modeling were reflected in the degradation model formula. The degradation model for predicting the lower limit consists of a deterministic trend term that indicates the degradation trend and a random term that represents variation factors. The model is formulated so that the trend and random terms match the actual phenomena. A parametric model was used so that the model error does not become excessive in case of extrapolation conditions, even with a small amount of training data. Equation (1) shows the basic model formula.

$$dX(t) = \mu(\eta(t), \theta_0) d\eta(t) \quad (1)$$

where

$$\eta(t) = t^b$$

Here, $X(t)$ is the capacity degradation rate (1 - capacity retention rate (%)) at time t ; θ_0 is a parameter of the degradation speed; $\mu(t, \theta_0)$ is the partial derivative ($\mu(t, \theta_0) = \frac{\partial}{\partial t} \Lambda(t, \theta_0)$) of the trend term $\Lambda(t, \theta_0)$ with respect to t ; $\eta(t)$ is a function that converts the scale of time $t^{(12)}$; and b is the scale conversion factor.

The random term that represents variation is noted as the stochastic variable ϵ that follows a normal distribution with a mean of 0 and a variance of σ_ϵ^2 (both mean and variance constant over time) (Table 1).

For durability variation that expresses behavior that gradually expands over time, a diffusion process was adopted^{(11), (12)}. $W(t)$ in Table 1 is the standard Wiener process, and σ is the diffusion coefficient.

Model error is a variation term that accumulates in the trend term. This is noted as the stochastic variable λ that follows a normal distribution with a mean of μ_λ and variance of σ_λ^2 (both mean and variance constant over time) and accumulates in the trend term of the performance formula. Note that variables introduced for the purpose of replacing deterministic variables with stochastic variables in the model are generally referred to as random effects⁽¹³⁾. Also, the three random terms above are assumed to be independent of each other.

Table 2 shows multiple candidate models based on combinations of how the above variation terms are incorporated.

In this study, a total of $2^3-1=7$ types of model formulas were formulated based on the presence or absence of the three types of error. However, there is no data that includes only model error without measurement variation or degradation variation, so that case was excluded from the candidate models, and the six models M1 to M6 were examined. Note that M1 is the conventional model that considers only measurement error, which is equivalent to a model using the least squares method.

Table 1 Three types of degradation formulas with variation terms

Items	Independence of degradation trends	Time dependence of variation	Formula
Measurement variation	Yes	No	$X(t) = \Lambda(\eta(t), \theta_0) + \epsilon$ $\epsilon \sim N(0, \sigma_\epsilon^2)$
Interindividual variation	Yes	Yes	$dX(t) = \mu(\eta(t), \theta) d\eta(t) + \sigma_B dW(\eta(t))$ $W(\eta(t)) \sim N(0, \eta(t))$
Model variation	No	Yes	$dX(t) = \lambda \times \mu(\eta(t), \theta) d\eta(t)$ $\lambda \sim N(0, \sigma_\lambda^2)$

Table 2 Six types of model formulas with combinations of variation terms

Model No.	Measurement variation	Inter-individual variation	SIM variation	Model formula
M1	○			$X(t) = \Lambda(\eta(t), \theta_0) + \epsilon$
M2		○		$dX(t) = \mu(\eta(t), \theta_0) d\eta(t) + \sigma_B dW(\eta(t))$
M3		○	○	$dX(t) = \lambda \times \mu(\eta(t), \theta_0) d\eta(t) + \sigma_B dW(\eta(t))$
M4	○	○		$X(t) = Y(t) + \epsilon$ $dY(t) = \mu(\eta(t), \theta_0) d\eta(t) + \sigma_B dW(\eta(t))$
M5	○	○	○	$X(t) = Y(t) + \epsilon$ $dY(t) = \lambda \times \mu(\eta(t), \theta_0) d\eta(t) + \sigma_B dW(\eta(t))$
M6	○		○	$X(t) = Y(t) + \epsilon$ $dY(t) = \lambda \times \mu(\eta(t), \theta_0) d\eta(t)$

$$\eta(t) = t^b, t > 0, b > 0, \lambda \sim N(\mu_\lambda, \sigma_\lambda^2), \mu_\lambda > 0, \sigma_\lambda > 0, W(\eta(t)) \sim N(0, \eta(t)), \epsilon \sim N(0, \sigma_\epsilon^2), \sigma_\epsilon > 0$$

Table 3 List of trend model formulas

No.	Model	Formula	Number of coefficients
(i)	Linear	$\sum_{i=1}^5 a_i \times X_i + a_0$	6
(ii)	Linear + interaction	$\sum_{i=1}^5 a_i \times X_i + \sum_{1 \leq i < j \leq 5} a_{i,j} \times X_i \times X_j + a_0$	16
(iii)	Quadratic	$\sum_{i=1}^5 a_{i,i} \times X_i^2 + \sum_{i=1}^5 a_i \times X_i + a_0$	11
(iv)	Quadratic + interaction	$\sum_{i=2}^5 a_{i,i} \times X_i^2 + \sum_{i=1}^5 a_i \times X_i + \sum_{1 \leq i < j \leq 5} a_{i,j} \times X_i \times X_j + a_0$	21

$$X_1: \text{SOC}; X_2: \text{Temperature}; X_3: \Delta\text{SOC}; X_4: \text{I-charge}; X_5: \text{I-discharge}$$

The trend term is expressed as shown in Eq. (2) using the variable $a(\theta_0, t)$ that expresses the degradation speed.

$$\Lambda(\eta(t), \theta_0) = a(\theta_0, t) \times \eta(t) \quad (2)$$

$a(\theta_0, t)$ is a variable indicating the degradation speed composed of the load variables $\text{SOC}(t)$, $\text{Temperature}(t)$, $\Delta\text{SOC}(t)$, $\text{I-charge}(t)$, and $\text{I-discharge}(t)$ and parameters $(a_0, a_1, a_2 \cdots b)$. As shown in Table 3, the formulated models include linear models (i) and (ii) with first-order linear factor effects for each degradation factor, quadratic polynomial models (iii) and (iv) that do not correspond to first-order linear equations, and interaction models (ii) and (iv) that have interaction terms between factors. B uses a unique appropriate value depending on the type of battery⁽¹⁰⁾.

3. Construction of Parameter Optimization Process

3.1. Parameter Optimization Method

The trend term parameters ($a_0, a_1, a_2 \dots b$) and variation term parameters ($\sigma_\epsilon, \sigma_B, \sigma_\lambda, \mu_\lambda$) included in the model formula were optimized based on actual test data. As these include nonlinear parameters, parameter estimation was performed by the maximum likelihood method with the Log-Likelihood (LL) function as the object variable. Here, let $X_i(t_{ij})$ represent the stochastic process that expresses the performance value of cell i at the time t_{ij} . The LL function of model formula M5, which has the largest number of parameters among the proposed models M2 to M6, is given by Eq. (3). To help avoid getting stuck in local solutions in the optimization calculations, the maximum likelihood estimation (MLE) values were obtained using the Markov Chain Monte Carlo (MCMC) method^{(14), (15)}. Specifically, a normal distribution with large variance was set as the prior distribution of each parameter. The MCMC method employed the Hamiltonian Monte Carlo (HMC) method, which is known to work effectively even with a large number of parameters, and the MLE values were obtained by performing maximum a posteriori (MAP) estimation.

$$l(\theta) = -\frac{\sum_{i=1}^n m_i}{2} \ln(2\pi) - \frac{1}{2} \sum_{i=1}^n \ln|\Sigma_i| - \frac{1}{2} \sum_{i=1}^n (\Delta X_i - \mu_\lambda \Delta \Lambda_i(\theta_0))^T \Sigma_i^{-1} (\Delta X_i - \mu_\lambda \Delta \Lambda_i(\theta_0)) \quad (3)$$

However,

$$\{\theta = (\theta_0, b, \mu_\lambda, \sigma_\lambda^2, \sigma_B^2, \sigma_\epsilon^2),$$

$$\mu_\lambda \in R,$$

$$\sigma_\lambda > 0,$$

$$\sigma_B^2 > 0,$$

$$\sigma_\epsilon^2 > 0,$$

$$\Sigma_i = \sigma_\lambda^2 \Delta \Lambda_i(\theta_0) \Delta \Lambda_i(\theta_0)^T + \Omega_i,$$

$$\Omega_i = \sigma_B^2 \times \text{diag}(\Delta \eta_i) + \sigma_\epsilon^2 F,$$

$$F = \begin{pmatrix} 1 & -1 & 0 & 0 & 0 & \dots & 0 \\ -1 & 2 & -1 & 0 & 0 & \dots & 0 \\ 0 & -1 & 2 & -1 & 0 & \dots & 0 \\ 0 & 0 & -1 & 2 & -1 & \dots & 0 \\ 0 & 0 & 0 & -1 & 2 & \dots & 0 \\ \vdots & \vdots & \vdots & \ddots & \ddots & \ddots & \vdots \\ 0 & 0 & 0 & \dots & 0 & -1 & 2 \end{pmatrix}$$

$$\Delta X = (\Delta X_1, \dots, \Delta X_n)^T,$$

$$\Delta X_i = (\Delta X_{i,1}, \dots, \Delta X_{i,m_i})^T,$$

$$\Delta X_{i,j} = (X_i(t_{i,j}) - X_i(t_{i,j-1})),$$

$$X_i(t_{i,j}) : \text{amount of degradation of cell } i \text{ at time } t_{i,j}$$

$$\Delta \Lambda_i(\theta_0) = (\Delta \Lambda_{i,1}(\theta_0), \dots, \Delta \Lambda_{i,m_i}(\theta_0))',$$

$$\Delta \Lambda_{i,j}(\theta_0) = a(\theta_0, t_{i,j}) \{\eta(t_{i,j}) - \eta(t_{i,j-1})\},$$

$$a(\theta_0, t_{i,j}) : \text{degradation acceleration factor of cell } i \text{ at time } t_{i,j}$$

$$\Delta \eta_i = (\Delta \eta_{i,1}, \dots, \Delta \eta_{i,m_i})',$$

$$\Delta \eta_{i,j} = \eta(t_{i,j}) - \eta(t_{i,j-1}) = t_{i,j}^b - t_{i,j-1}^b$$

$$t_{i,j} : \text{time at which the amount of degradation of cell } i \text{ was observed for the } j\text{th time.}$$

$$1 \leq j \leq m_i, 1 \leq i \leq j \}$$

3.2. Model Formula Selection Method

Selection of the trend formula and variation model formula is determined by Akaike's Information Criterion (AIC) based on the training data. AIC is commonly used as an index enabling selection of the optimal model while helping to prevent overfitting to only the training data. AIC is calculated using Eq. (4) from the final LL value used as the object variable in optimization and the number of parameters in the model (k).

$$AIC = -2LL + 2k \quad (4)$$

k: number of parameters

3.3. Prediction of Lower Limit Performance

The distribution of $\Delta X_{i,1:m_i+1}$ under the given $\Delta X_{i,1:m_i}$ is obtained by the following Eq. (5).

$$\Delta X_{i, m_i+1} | \Delta X_{i, 1:m_i} \sim N(\mu_{m_i+1|m_i}, \sigma_{m_i+1|m_i}^2), \quad (5)$$

However,

$$\{\mu_{m_i+1|m_i} = \mu_\lambda \Delta \Lambda_{i, m_i+1}(\theta_0) + \omega'_{i,1} \Sigma_{i,1:m_i}^{-1} (\Delta X_{i,1:m_i} - \mu_\lambda \Delta \Lambda_{i,1:m_i}(\theta_0))$$

$$\sigma_{m_i+1|m_i}^2 = \sigma_{i, m_i+1}^2 - \omega'_{i,1} \Sigma_{i,1:m_i}^{-1} \omega_{i,1},$$

$$\omega_i = (0, \dots, 0, -\sigma_\epsilon^2)', \quad (m_i\text{-dimensional vector}),$$

$$\Sigma_{i,1:m_i} = \sigma_\lambda^2 \Delta \Lambda_{i,1:m_i}(\theta_0) \Delta \Lambda_{i,1:m_i}(\theta_0)' + \Omega_{i,1:m_i},$$

$$\Omega_{i,1:m_i} = \sigma_B^2 \text{diag}(\Delta \eta_{i,1:m_i}) + \sigma_\epsilon^2 F_{obs},$$

$$F_{obs} = \begin{pmatrix} 1 & -1 & 0 & 0 & 0 & \dots & 0 \\ -1 & 2 & -1 & 0 & 0 & \dots & 0 \\ 0 & -1 & 2 & -1 & 0 & \dots & 0 \\ 0 & 0 & -1 & 2 & -1 & \dots & 0 \\ 0 & 0 & 0 & -1 & 2 & \dots & 0 \\ \vdots & \vdots & \vdots & \ddots & \ddots & \ddots & \vdots \\ 0 & 0 & 0 & \dots & 0 & -1 & 2 \end{pmatrix},$$

($m_i \times m_i$ matrix),

$$\Sigma_i^{-1} = \Omega_{i,1:m_i}^{-1} - \frac{\sigma_\lambda^2}{1 + \sigma_\lambda^2 \Delta \Lambda_{i,1:m_i}(\theta_0)' \Omega_{i,1:m_i}^{-1} \Delta \Lambda_{i,1:m_i}(\theta_0)} \times \Omega_{i,1:m_i}^{-1} \Delta \Lambda_{i,1:m_i}(\theta_0) \Delta \Lambda_{i,1:m_i}(\theta_0)' \Omega_{i,1:m_i}^{-1},$$

$$\sigma_{i, m_i+1}^2 = \sigma_\lambda^2 \Delta \Lambda_{i, m_i+1}(\theta_0)^2 + \sigma_B^2 \Delta \eta_{i, m_i+1} + 2\sigma_\epsilon^2 \}$$

Therefore, when the upper $\alpha\%$ point of the standard normal distribution is noted as z_α , the $100 \times (1-\alpha)\%$ confidence interval for the degradation increment ΔX_{ij} of the individual cell number i at time t_{i,m_i+1} , under the given $\Delta X_{i,1:m_i}$, is given by Eq. (6).

$$\left[\hat{\mu}_{m_i+1|m_i} - \hat{\sigma}_{m_i+1|m_i} z_\alpha, \hat{\mu}_{m_i+1|m_i} + \hat{\sigma}_{m_i+1|m_i} z_\alpha \right] \quad (6)$$

3.4. Preparation of Training and Validation Data

The results of durability tests conducted under multiple conditions were used as the data set. The tests utilized 4.5 Ah-class cylindrical LiBs as test samples, and were conducted in two modes: storage durability tests and cycle durability tests. The storage durability tests for training data were conducted over a 4-month period under 16 conditions comprising all combinations of four SOC conditions (SOC30, 55, 80, and 95%) and four temperature conditions (25, 45, 50, and 65°C). During the testing period, intermediate performance measurements were conducted at intervals of one to two weeks, and 10 data points were obtained per condition for a total of 160 data points. The cycle durability tests were conducted under 27 conditions planned based on an L27 orthogonal array comprising five cycling control factors: center SOC, operating SOC range, temperature, charge current, and discharge current. Ten data points were obtained over a 4-month period for each condition, for a total of 270 data points. For the validation data, durability tests were conducted over a period of 12 months under 38 conditions separate from the training conditions, and obtained a total of 710 data points.

4. Results and Discussion

4.1. Model Selection Result

Regarding model selection, parameter optimization was performed for 24 types of model formulas comprising combinations of four types of trend term model formulas and six types of variation models (M1 to M6), and the AIC was evaluated. Figure 4 shows the results.

In all cases of M2 to M6 constructed in this study, good results were obtained with an AIC lower than that of the conventional model M1. This demonstrates that model formulas using the diffusion process, random effects, or both can appropriately represent the experimental data.

Generally, the accuracy of the model formula increases with the number of parameters, but too many parameters lead to a drop in generalization performance. Therefore, it is appropriate to select the model with the smallest AIC that also balances generalization performance and goodness of fit. Of the degradation model formulas, M3 has fewer parameters than M5 but also a smaller AIC. That is to say, M3 captures the essence of the battery performance degradation obtained from the tests with fewer parameters. Here, the combination of the quadratic (iii) trend term formula and the M3 model formula resulted in the best fit to

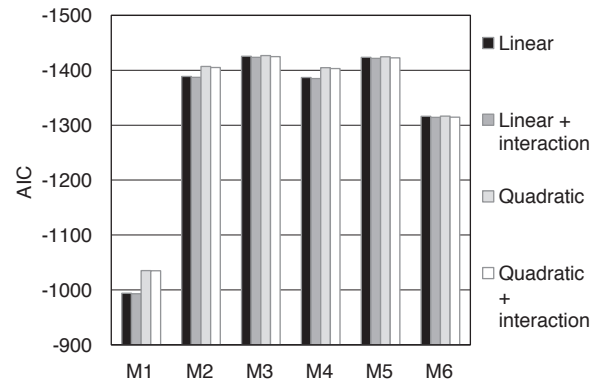
the experimental data. Also, it was found that enhancement of AIC was not observed with models M4 to M6, which include measurement variation, so there is no need to consider these in the model.

4.2. Prediction of Confidence Interval of Selected Model

Equation (7) shows the model formula and parameters of the prediction model M3 selected in the previous section. In addition, Fig. 5 shows plots of the measurement results from a 12-month durability test conducted at SOC100% and temperature of 45°C, along with an example of the confidence interval prediction result calculated using the optimized M3 degradation model. The confidence interval indicated in gray is the 95% confidence interval at each point in time. The measured plots across the entire range were confirmed to fall within the 95% confidence interval, thus demonstrating the ability to appropriately visualize the confidence interval.

$$\begin{aligned} dX(t) &= \lambda \times \mu(\eta(t), \theta_0) d\eta(t) + \sigma_B dW(\eta(t)) \\ \mu(\eta(t), \theta_0) d\eta(t) &= (a_1 SOC + a_2 Temp + a_{11} SOC^2 + a_{22} Temp^2 + a_0) d\eta(t) \\ \eta(t) &= t^b \end{aligned} \quad (7)$$

Term	Value	Term	Value
a_0	-5.13	μ_λ	-1.7E-2
a_1	2.6	σ_λ	1.6E-3
a_2	-1.7E-3	σ_B	7.3E-4
a_{11}	-3.2E-1		
a_{22}	1.0E-4		
b	0.55		



		Variation models					
	Parameter	1	2	3	2	4	3
	Parameter	M1	M2	M3	M4	M5	M6
Model formula of trend term	4 (i) Linear	-993.8	-1388.7	-1425.5	-1386.7	-1423.5	-1316.3
	5 (ii) Linear+ interaction	-992.8	-1387	-1423.5	-1385	-1421.5	-1314.3
	6 (iii) Quadratic	-1035.3	-1406.8	-1426.7	-1404.8	-1424.6	-1316.6
	7 (iv) Quadratic+ interaction	-1034.8	-1405.1	-1424.7	-1403.1	-1422.6	-1314.7

Fig. 4 AIC results of each model

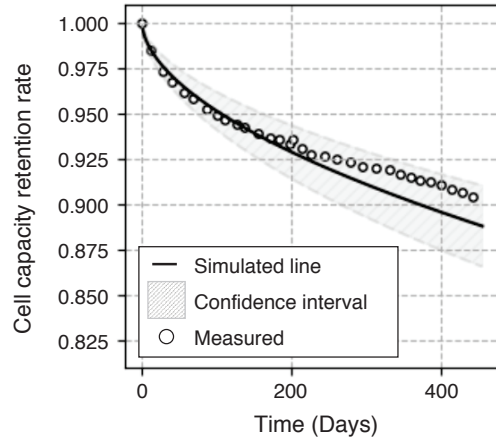
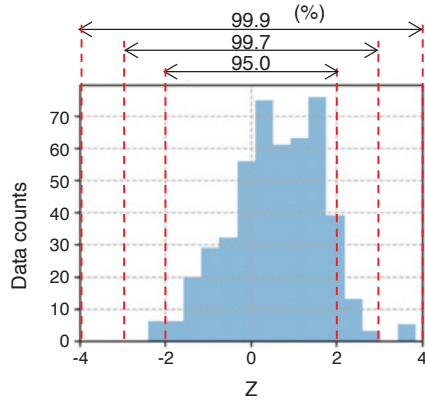


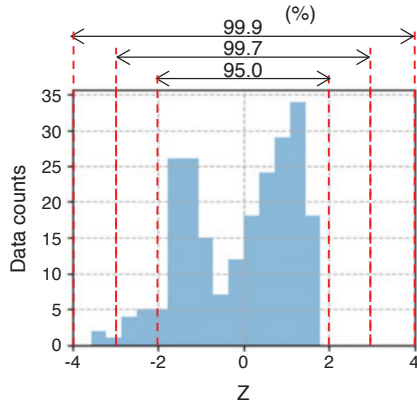
Fig. 5 Confidence interval and measured data points

4.3. Confirmation of Data Distribution

To confirm the likelihood of the model created in this study, the degree of coverage of the predicted performance distribution relative to the measured performance values was analyzed for an amount of data greater than the training data (storage degradation mode data: 24 conditions, 484 points; cycle degradation mode data: 14 conditions, 226 points). Figure 6 shows histograms of the Z values



(a) Histogram of data obtained by storage mode degradation



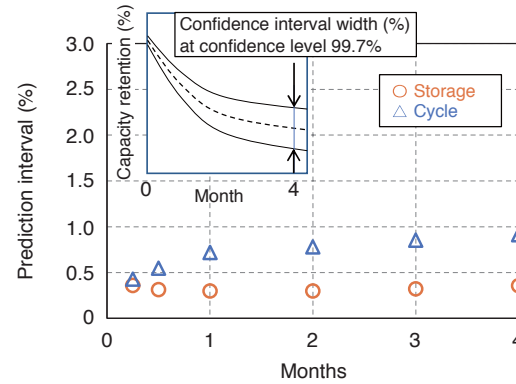
(b) Histogram of data obtained by cycle mode degradation

Fig. 6 Histograms of Z values after standardization

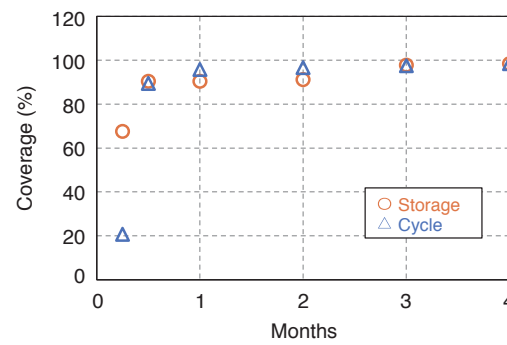
obtained by taking the difference between the typical performance value predicted by the model and the measured value and dividing by the standard deviation at that point. Figure 6(a) is the histogram of the data obtained in storage degradation mode, and Fig. 6(b) is the histogram of the data obtained in cycle degradation mode. The Z values of 4, 3, and 2 correspond respectively to the confidence intervals that theoretically contain 99.9%, 99.7%, and 95%. In this study, these confidence intervals respectively contained 100%, 99.0%, and 93.6% of the storage mode data and 100%, 98.7%, and 94.0% of the cycle mode data, generally matching the theoretical values. It is thought, therefore, that lower limit performance based on confidence can be calculated using this model. The percentage of the measured data contained in the 99.7% confidence interval is referred to herein after as the coverage percentage.

4.4. Setting of Training Period

In order to set the period required for model construction, the amount of training data, the width of the confidence interval, and the coverage percentage were analyzed. The width of the confidence interval (%) and the coverage percentage in the 99.7% confidence interval predicted by each model were calculated at the 4-month point, with Fig. 7 showing the results.



(a) Confidence interval width



(b) Coverage

Fig. 7 Confidence interval width and coverage of models trained with data over different periods

A model was constructed using the data up to an arbitrary number of months from four months of training data obtained by storage durability tests under 16 conditions and cycle durability tests under 27 conditions. The coverage percentage was calculated for a total of 380 data points obtained under 38 conditions separate from the training data.

The width of the confidence interval tends to increase slightly as the amount of training data increases. In contrast, the coverage percentage clearly increases, and the coverage percentage of the 99.7% confidence interval increases asymptotically toward 99.7% as theorized. This is presumed to be due to the effect that as the amount of training data increases, the accuracy of the predicted confidence interval is enhanced without greatly impacting the confidence interval result. In this study, a coverage percentage of 99% or more was achieved with a 4-month training period, indicating that the confidence interval can be predicted by training on four or more months of degradation behavior data.

4.5. Confidence Interval in Long-term Prediction

Prediction of the performance degradation of batteries that are used in various ways becomes more uncertain as the prediction period lengthens, which is expected to result in a wider confidence interval.

Figure 8 shows the results of long-term prediction of the confidence interval over 10 years. Figure 8(a) shows the result for the conventional model M1 that considers only measurement variation, and Fig. 8(b) shows the result for the newly proposed optimized prediction model M3.

Whereas the conventional model predicts a uniform confidence interval over a period of 10 years, the model optimized in this study exhibits a confidence interval that expands over time, which is in line with initial expectations. It was confirmed that, compared to the conventional model M1, the model M3 proposed in this study can appropriately model the increase over time of variation appearing in four months of test data.

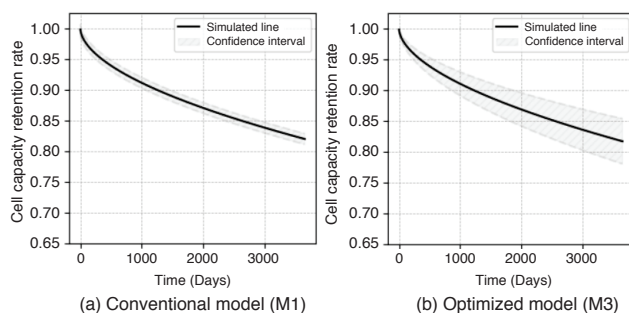


Fig. 8 Simulated confidence interval for 10 years

5. Conclusion

In order to use a model to efficiently predict the lower limit of lithium-ion battery performance after durability

testing, a degradation model formula that incorporates random terms was examined. The conclusion drawn from this examination is as follows.

- (1) It was found that the variation in lithium-ion battery durability data can be represented by a model incorporating two kinds of stochastic variables: random effects multiplied by the LiB durability degradation trend term, and a diffusion process in which variation expands proportionally with time.
- (2) Use of the MLE method aided by MCMC enables optimization of the model without getting stuck in local solutions.
- (3) A model was constructed that can appropriately predict the confidence interval over a 12-month period by training on only four months of durability test results.

References

- (1) New Energy and Industrial Technology Development Organization: Battery Road Map 2013, (2013) (in Japanese), https://www.jstage.jst.go.jp/article/ieejias/134/3/134_NL3_5/_pdf, (accessed 2024/12/24)
- (2) Fuji Keizai Group Co., Ltd.: Energi, ogata nijidenchi, zairyo no shorai tenbo 2021 - Dendo jidosha, shasaidenchi bunya hen -, p. 5-12, (2021) (in Japanese)
- (3) METI: Ministry of Economy, Trade and Industry, https://www.meti.go.jp/policy/mono_info_service/joho/conference/battery_strategy/0001/03r2.pdf, (accessed 2024/12/24) (in Japanese)
- (4) Dunn, B., Kamath, H., Tarascon, J.-M.: Electrical Energy storage for the Grid: A Battery of Choices, *Science*, Vol. 334, Issue 6058, p. 928-935, (2011), doi: 10.1126/science.1212741
- (5) Schmuck, R., Wagner, R., Hörpel, G., Placke, T., Winter, M.: Performance and cost of materials for lithium-based rechargeable automotive batteries, *Nature Energy*, Vol. 3, p. 267-278, (2018), doi: 10.1038/s41560-018-0107-2
- (6) Final Regulation Order, Section 1962.8, Title 13, California Code of Regulations, (2022), <https://www2.arb.ca.gov/sites/default/files/barcu/regact/2022/accii/accii1962.8.pdf>, (accessed 2024/12/24)
- (7) Waag, W., Fleischer, C., Sauer, D. U.: Critical review of the methods for monitoring of lithium-ion batteries in electric and hybrid vehicles, *Journal of Power Sources*, Vol. 258, p. 321-339, (2014), doi: 10.1016/j.jpowsour.2014.02.064
- (8) McGrasso, J., Ressler, G., Inguva, S.: MEETING CUSTOMER EXPECTATIONS: Life and Safety of Batteries in Hybrid and Electric Vehicles, SAE Technical Paper TP-AAB-AABC_US_2011_20, (2011)
- (9) Tominaga, Y.: Degradation Model for Battery Performance Based on Performance Mechanism, *The Electrochemical Society of Japan*, Vol. 89, No. 2, p.

- 125-132, (2021), doi: 10.5796/denkikagaku.21-FE0014
(in Japanese)
- (10) Omichi, K., Kaga, T., Nishiimura, T., Aida, N., Tominaga, Y.: Modeling of Efficient Lifetime Prediction for Lithium-ion Battery Employing Orthogonal Array, Honda R&D Technical Review, Vol. 35, p. 32-38
 - (11) Xu, X., Tang, S., Yu, C., Xie, J., Han, X., Ouyang, M.: Remaining Useful Life Prediction of Lithium-ion Batteries Based on Wiener Process Under Time-Varying Temperature Condition, Reliability Engineering & System Safety, Vol. 214, (2021), doi: 10.1016/j.ress.2021.107675
 - (12) Russell, M. B., King, E. M., Parrish, C. A., Wang, P.: Stochastic modeling for tracking and prediction of gradual and transient battery performance degradation, Journal of Manufacturing Systems, Vol. 59, p. 663-674, (2021), doi: 10.1016/j.jmsy.2021.04.008
 - (13) Wu, C. F. J., Hamada, M. S.: Experiments: Planning, Analysis, and Optimization, John Wiley & Sons, Inc., (2021), doi: 10.1002/9781119470007
 - (14) Castillo, E., Hadi, A. S.: Modeling Lifetime Data with Application to Fatigue Models, Journal of the American Statistical Association, Vol. 90, Issue 431, p. 1041-1054, (1995), doi: 10.2307/2291341
 - (15) Jacquier, E., Johannes, M., Polson, N.: MCMC maximum likelihood for latent state models, Journal of Econometrics, Vol. 137, Issue 2, p. 615-640, (2007), doi: 10.1016/j.jeconom.2005.11.017

■ Author ■



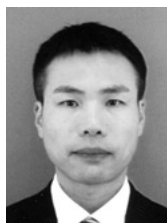
Kaoru OMICHI



Kotone SUZUKI



Hideki NAGATSUKA



Michihiro YASUDA