Battery Degradation Modeling Based on FIB-SEM Image Features Extracted by Deep Neural Network

Dr. Yoichi TAKAGISHI*1 • Dr. Tatsuya YAMAUE*1
*1 Kobelco Research Institute, Inc

Abstract

Attention is being paid to attempts at predicting the degradation and life of lithium-ion batteries (LIBs). This paper focuses on the examination conducted on the features, advantages, disadvantages, etc., of a data-driven prediction model that combines feature extraction and regression by deep learning. Also described is a physics-based model that predicts the degradation progress by electrochemical reaction formula and the like. As a result, it was found that in the physics-based model, the prediction accuracy is high when the degradation phenomena are relatively straightforward, but its application is difficult when the phenomena are complicated or unknown. On the other hand, the data-driven modeling can be done even when the phenomena are not sufficiently clear and is considered to have a great advantage in predicting degradation accurately. Further consideration of the constructed model has also turned out to be useful for elucidating hidden phenomena.

Introduction

Lithium-ion batteries (hereinafter referred to as "LIBs") are installed in various equipment, including information terminals such as notebook PCs and smartphones, as well as transport machines such as electric cars, aircraft, and artificial satellites. Therefore, the technologies for predicting the degradation of LIBs have become extremely important in evaluating equipment life.

The degradation prediction methods for LIBs are being actively studied, including, in addition to the simple rule of thumb (power law, logarithmic law), physics-based models (electrochemical models) in which hypotheses of degradation phenomena are human-made to solve electrochemical reaction formulas numerically, and, more recently, data-driven models for predicting the remaining life from charge-discharge cycle data based on machine learning. **Fig. 1** is a map summarizing these approaches, showing "Hypothesis-driven/Data-driven" on the vertical axis and "Black box/White box" on the horizontal axis. In the hypothesis-driven approaches, various assumptions (simplification or homogenization) are human-made to model the phenomena. In contrast, in the "data-driven" approaches, models that conform to the measured data are established by machine learning. The term "black box" refers to a modeling method of describing a phenomenon only by the relationship between input and output, and "white box" refers to a modeling method that takes into account the details of the phenomenon (here, electrochemical reaction, concentration diffusion, and the like). The empirical methods (second quadrant) as typified by route rules[1] are easy to apply because they use simple functions, but their validity must always be discussed. Physics-based models[2] (first quadrant) are rigorous because they assume physical phenomena. Still, it is challenging to establish a model when the phenomenon itself is complicated or if there is an unknown process.

Recently, the data-driven approach[3] (third quadrant) has been attracting attention. In this approach, machine learning is proactively applied to degradation data under various conditions. Most of these attempts, however, are black-box types not considering the mechanism. Although they show excellent regression/prediction performance, feedback on the cell design and operating conditions is difficult because degradation factors are often hard to estimate.

KOBELCO Research Institute Inc. has recently proposed a white box method that assumes degradation phenomena while taking a data-driven approach (4th quadrant).[4] This paper briefly describes the conventional physics-based model, which models the complex phenomena of degradation, and then describes the data-driven white box model that predicts degradation from the SEM images of electrode cross-sections.

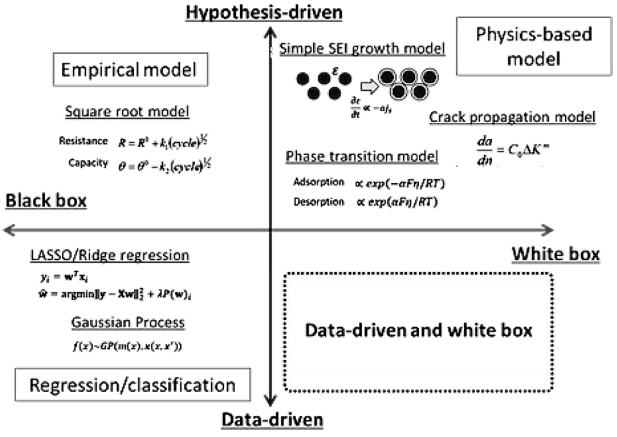

Fig. 1 Approach map for predicting degradation of Li-ion batteries

1. Conventional physics-based model

A method (physics-based model) that considers various degradation phenomena inside electrodes in ordinary or partial differential equations has been suggested as a high-precision method of predicting LIB degradation behavior.[1] **Fig. 2** shows an example of the degradation prediction schemes using a physics-based model. First, various observations, such as SEM and TEM/EELS, are performed to identify critical phenomena in battery degradation. In the case of LIBs, the main degradation phenomena include passivation films (solid electrolyte interfaces, hereinafter referred to as "SEIs") deposited on the active material surface, structural transition in the surface layer of the active material, and inter-particle cracks. Modeling methods such as the "simple SEI growth model," "phase transition model," and "crack propagation model" are suggested, respectively. Next, the formula predicting the temporal evolution of the physical values is established for the identified degradation phenomenon. For example, in the case of SEI film growth, the side reaction current is usually calculated using Tafel's equation. It is often assumed that the growth rate of film thickness is proportional to this current. Finally, the physical values are reflected and converted into battery characteristics (charge-discharge resistance and capacity) to predict the subsequent behavior. The parameters used for the case (hereinafter referred to as "reflection parameters") are determined by fitting them to the actual measurement. The fitting includes sequential optimization by the least square method with regularization term and data assimilation by Kalman filter. **Fig. 3** shows an example of applying the physics-based model to the degradation prediction of a $Li(Ni_{1/3}Mn_{1/3}Co_{1/3})O_2$-graphite system, a typical configuration of LIB electrodes. A first-order reaction formula expresses the coverage increase for the structural transition layer of the positive electrode active material. Two types of negative electrode SEI films (inorganic and organic) are assumed, and the linearized Butler-Volmer equation is used for the growth rate, in which the growth driving force is regarded to be proportional to the side reaction overvoltage. In addition, the crack growth rate between active material particles or at the interface of the current collector foil adopts an empirical formula based on the Paris rules.

Such an approach can be expected to result in a highly accurate prediction compared with the empirical rule of thumb[2] but requires the assumptions of degradation factors in advance. Therefore, the key is to model the dominant factor properly. Especially, modeling itself may be difficult if the degradation phenomena are complex or the factors are not sufficiently clear.

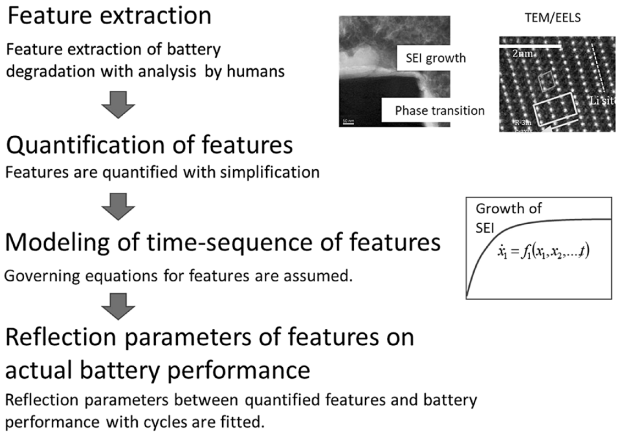

Fig. 2 Typical degradation modeling methods using physics-based models

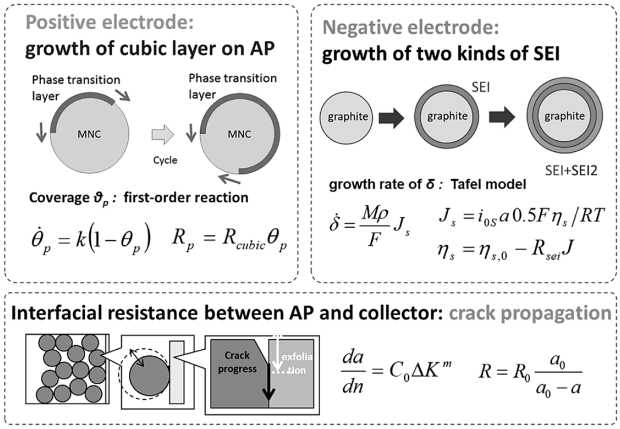

Fig. 3 Typical physics-based model in Li ($Ni_{1/3}Mn_{1/3}Co_{1/3}$) O_2—graphite system

2. Data-driven white box model

KOBELCO Research Institute Inc. has recently developed a new degradation prediction technology. This technology comprises extracting statistical feature values by deep learning and image analysis from the cross-sectional SEM image of each degraded state of the electrode and estimating their temporal evolution by the Gaussian process regression. This technology's significant advantage is that the degraded state's feature value is extracted and selected from analysis images, which enables a highly accurate prediction while estimating the objective degradation factors.

Fig. 4 shows the flow of the analysis. As described in the previous section, a data-driven model may be established using only the charge-discharge voltage's cycle/temporal change data. However, despite being of the same data-driven

type, the present approach uses SEM images of the electrode as the input value. It thus is a white box approach expected to enable the feature acquisition of degradation phenomena inside the battery. The main flow is as follows:

① Feature value is extracted by deep learning and visualized in the analysis images (here, the SEM images of the electrode cross-section) of each degraded state to identify the notable feature locations in the degradation image. In addition, image processing is used to score the statistical feature value of degradation indicated by these feature locations. As will be described later, scoring candidates include the area, thickness, and maximum pore diameter of the coated and/or cracked regions.

② A nonparametric regression without prescribing a function form is used to predict how these scores change over time.

③ An artificial neural network determines the reflection parameters that map each score to the actual battery capacity degradation value.

2.1 Acquisition of measurement data

A battery cell for evaluation (prototype coin cell) was created to acquire measurement data for machine learning. The electrodes and electrolyte were made of ordinary materials as much as possible, adopting Li $(Ni_{1/3}Co_{1/3}Mn_{1/3})O_2$ for the positive electrode, SiO-graphite for the negative electrode, and 1M $LiPF_6$(EC: DEC= 1 : 1) for the electrolytes. The coin cell had a diameter of 10 mm and a capacity of 4.8 mAh.

Next, after the initial charge-discharge, a charge-discharge cycle test was performed under a temperature of 25 ℃, an upper limit voltage of 4.0 V, a lower limit voltage of 3.0 V, and a charge-discharge rate of 1 C to measure the capacity per each cycle. In addition, the cross-section of the negative SiO electrode in each degraded state was observed by SEM. The maximum number of cycles was set to 100.

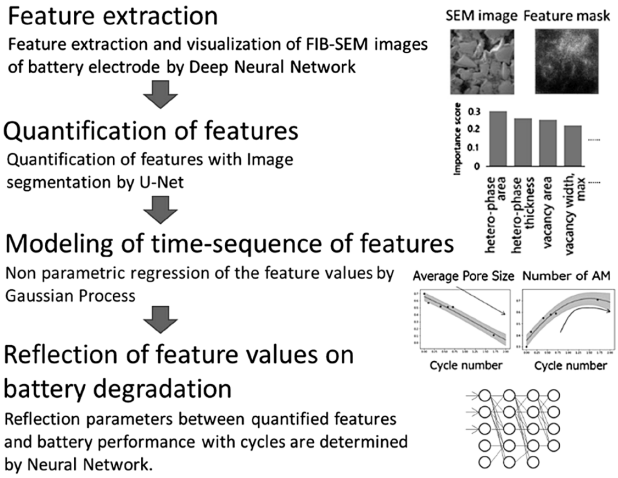

Fig. 4 Proposed procedure for data-driven and white box approaches

2.2 Extraction and selection of feature values from FIB-SEM image

The cross-sectional SEM images of negative electrodes in the initial state and degraded state (after ten cycles) were trimmed and enlarged to 50 μm squares to establish a degradation state classification model using a convolutional neural network (hereinafter referred to as "CNN"). The CNN feature value extractor was based on VGG16,[5] having a proven track record in ImageNet 1000 class classification. The weight of the neural network was adjusted independently only for the fully connected layer and the previous block. The 4,096-dimensional feature value vector thus obtained was used to carry out classification. As a result, learning approximately 50 images yielded excellent classification characteristics. These flows are shown in **Fig. 5**. Furthermore, the slope of the loss function was backpropagated to the input image to visualize the pixels contributing to the classification.[6]

The mask image, thus obtained, indicates high reactions in relatively vast pores, precipitation films around the active material, and the fine porous region (**Fig. 6**), which leads to an interpretation that these are the regions characterizing the degradation. It should be noted, however, that the vectors representing these degradation characteristics are not interpretable as-is and cannot necessarily be used for degradation prediction. Therefore, this study used U-net,[7] a segmentation method based on deep learning. This technique has been used to perform the regional division for each degraded

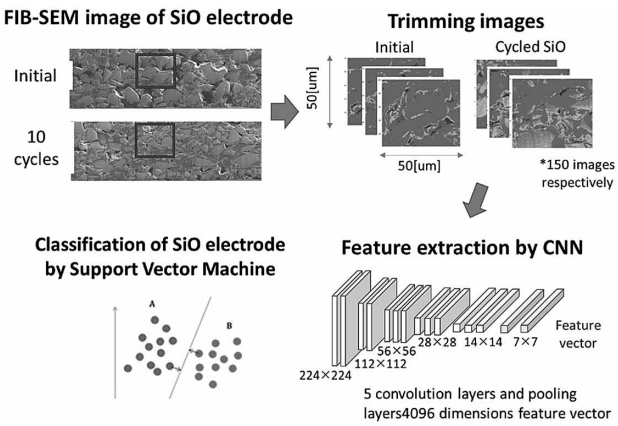

Fig. 5 Flow of feature extraction and classification of FIB-SEM images using deep learning

state image to identify the precipitation film and porous regions found in the feature locations of the cross-sectional SEM image. At this point, annotation by a skilled SEM technician was carried out to create teacher data. Statistical values on the image such as pore area, surface roughness, and average film thickness of the active material surface layer were calculated and defined as the degradation feature values. Here, segmentation was carried out in four regions: active material, binder/conductive aid, pores, and active material surface layer (**Fig. 7**).

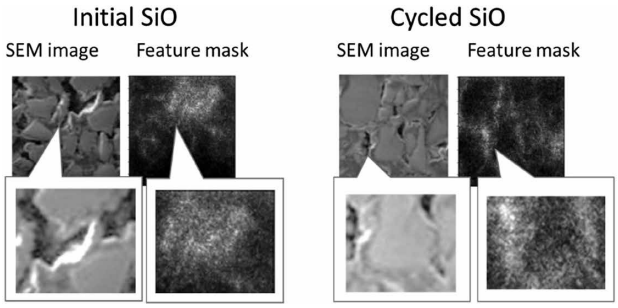

Fig. 6 Visualization of feature locations in cross-sectional SEM images of initial (left) and degraded (right) SiO_x electrodes

2.3 Cycle change of degradation feature values

In Section 2.2, the degradation behavior of battery capacity has been predicted by Gaussian process regression with the number of charge-discharge cycles as the parameter for the temporal change of the quantified degradation feature values. The Gaussian process regression is one of the nonparametric regression methods. It comprises probabilistically interpolating between the data points by the kernel function, thereby adaptively performing regression and prediction from the data characteristics without specifying the function shape. In the present analysis, the Gaussian kernel given by Eq. (1) has been adopted:

$$\kappa_{SE}(x, x') = \theta_f^2 \exp\left(-\frac{1}{\theta_l^2}\|x - x'\|^2\right) \quad \cdots\cdots\cdots\cdots\cdots (1)$$

wherein x and x' are data vectors, and θ_f and θ_l are scaling parameters.

Fig. 8 (figures on the left) shows an exemplary regression for the change of degradation feature values with the cycle. This example deals with the

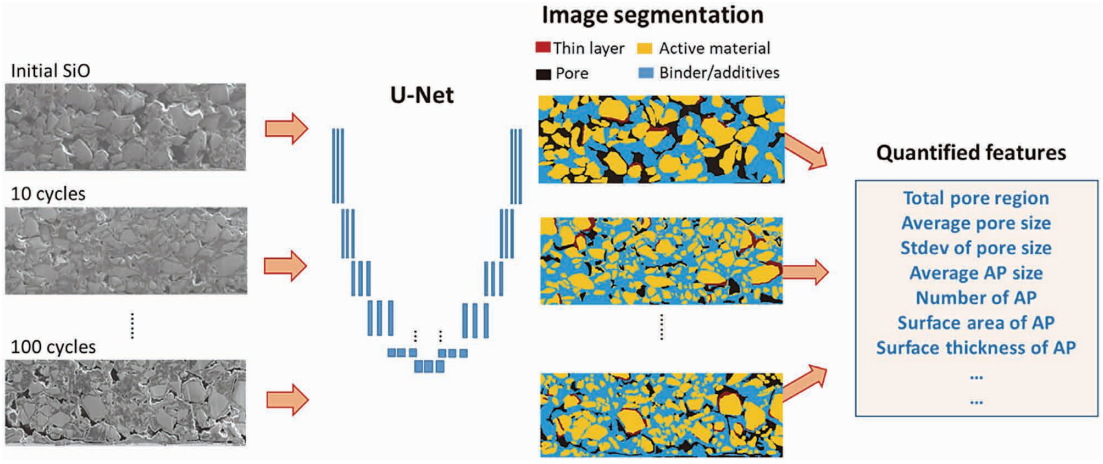

Fig. 7 Segmentation of FIB-SEM images of SiO_x using U-net

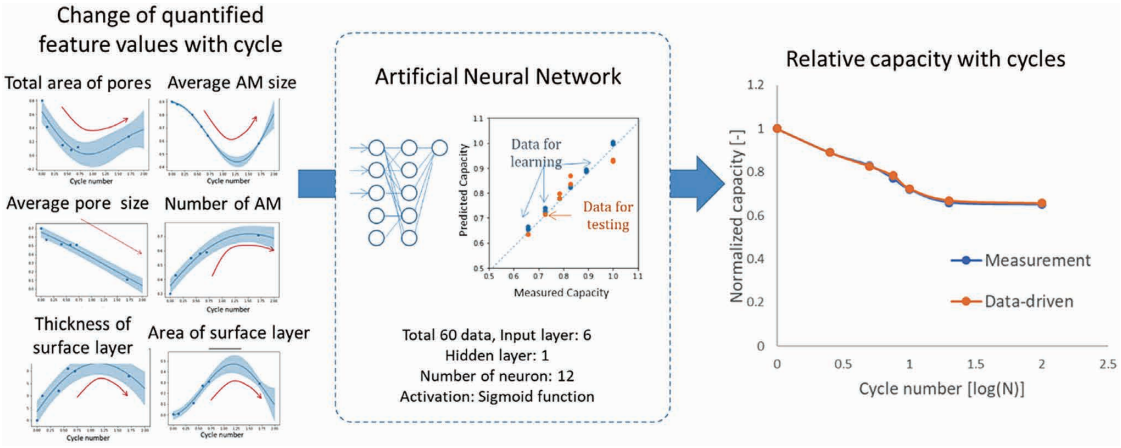

Fig. 8 Example of regression of cycle change in degradation features

cycle changes in the entire area of pores, the average area of the active material, the entire area and thickness of the surface region of the active material, the average pore size, and the number of active materials. While the measured battery capacity decreased monotonously, each score fluctuated with the cycle, exhibiting a complicated behavior. For example, the thickness of the active material surface layer does not necessarily increase monotonously with the cycle. In other words, the conventional physics-based model assumes relatively monotonous temporal changes in physical values, but does not necessarily capture the phenomena sufficiently.

The combination of those degradation feature values, subjected to regression, is considered to correspond with the value of electrode degradation. Therefore, the cycle change of each score and the measured degradation value (capacity drop) have been coordinated with the reflection parameters (Fig. 8, center figure). In the present analysis, the coordination has been conducted by an artificial neural network with each degradation feature value as an input and reflection parameter as an output. It should be noted that the reflection parameters have been assumed to have no dependence on the number of cycles. Since the capacity of actual measurement limits the number of data points, the regression has been performed after interpolating the data. Fig. 8 (figure to the right) compares the measured capacity retention rate and the capacity retention rate determined by the regression of the degradation values. The model demonstrates an excellent regression.

2.4 Considerations

The contribution of each degradation feature value has been analyzed from the input layer and the coefficient of the middle-class first layer of the established neural network. **Fig. 9** (a) shows the contribution of each factor to the capacity loss after each cycle. Higher values are indicated by the feature values related to the active material size, such as the average area of active material and the active material density, and by the pore diameter, suggesting their significant contribution. Fig. 9 (b) shows the segmentation images of the negative electrode cross-section after the initial 10 cycles and 100 cycles. As shown, the active material size decreases, and the finer pores increase as the cycle progress. Thus, the degradation is presumed to be mainly due to the pulverization of active material and crack propagation, which enables feedback to the electrode design.

As described, this approach uses the SEM images of the electrode as input and enables degradation prediction for phenomena whose mechanism is not sufficiently clear. It is also noteworthy that the features related to electrode degradation and the main factors have been extracted.

The present analysis uses the cross-sectional SEM images as the only learning data. The model is expected to be further enhanced by including other information (including XPS and TEM-EELS). On the other hand, this approach assumes no physical phenomenon, making the data quality extremely important. The challenge is to secure a sufficient quality and amount of data. In addition, the relationship between the feature values extracted here is nothing more than an inter-phase

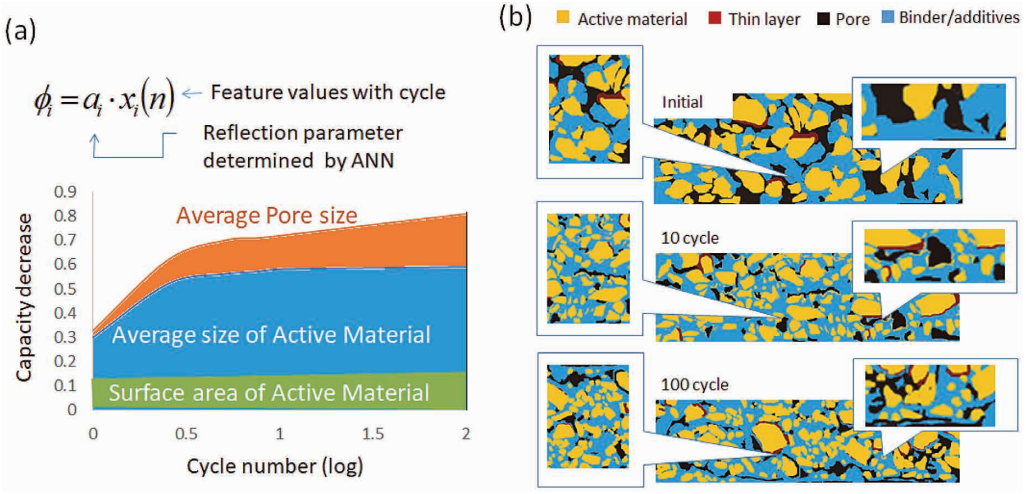

Fig. 9 Cycle change of parameters reflected in actual measurement (a), and segmented FIB-SEM images at initial cycle, after 10 cycles, and after 100 cycles (b)

relationship, and it is necessary to pay sufficient attention to spurious correlation.

Conclusions

This paper has described the degradation prediction technology of lithium-ion batteries and introduced a conventional physics-based model and a data-driven model using machine learning and deep learning, attracting recent attention. The newly developed data-driven white box models enable the modeling of insufficiently clear phenomena and have highly superior degradation prediction accuracy. It has been shown that considering an established model is also helpful in elucidating hidden phenomena. Each of these models has its own character, and it is essential to use them properly in accordance with the phenomena, purpose and findings.

The plan includes establishing highly accurate prediction technology, including extrapolation region by utilizing the SINDy method[8] that estimates the governing equation from data by sparse matrix.

References

1) G. Ning et al. Journal of Power Sources. 2003, Vol.117, pp.160-169.
2) Y. Takagishi et al. International Journal of Automotive Engineering. 2017, Vol.8, No.3, pp.143-148.
3) R. Richardson et al. Journal of Power Sources. 2017, Vol.357, pp.209-219.
4) Y. Takagishi et al. 59th Battery Symposium in Japan. 2018-11-27/29, The Electrochemical Society of Japan, p.209.
5) D. Smilkov et al. SmoothGrad: removing noise by adding noise. arXiv.org e-print archive. 2017, 1706.03825, https://arxiv.org/abs/1706.03825, (accessed on 2020-12-14).
6) K. Simonyan et al. Very Deep Convolutional Networks for Large-Scale Image Recognition. arXiv.org e-print archive. 2015, 1409.1556. https://arxiv.org/abs/1409.1556, (accessed on 2020-12-14).
7) O. Roneberger et al. U-Net: Convolutional Networks for Biomedical Image Segmentation. arXiv.org e-print archive.2015, 1505.04597. https://arxiv.org/abs/1505.04597, (accessed on 2020-12-14).
8) S. L. Brunton. et al. Acad. Sci. U.S.A. 2016, Vol.113, pp.3932-3937.