

Semi-physical Model that Predicts Internal State of Lithium-ion Batteries and Calculates Degradation Behavior with High Accuracy

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ABSTRACT

A degradation simulation model based on the mechanism of lithium-ion rechargeable battery performance expression was developed in order to predict the long-term durability of on-board lithium-ion rechargeable batteries with high accuracy. Up to now, guaranteeing the long-term durability of lithium-ion rechargeable batteries required correspondingly long-term testing and design values with a large margin in terms of prediction accuracy. In the developed model, the model portion that calculates the battery performance was made into a physical model using the physical quantities that influence the degradation mechanism as explanatory variables, and the degradation behavior of each factor was made into an empirical model that is trained from the test results. The model with this hybrid structure was proposed as a semi-physical model. As a result of this, accuracy in predicting durability was increased, and a capability to represent the non-linear behavior of performance degradation transition over long periods of time was indicated. When the internal state of that model is to be acquired by disassembly analysis, however, model training incurs a high development cost. For this investigation, therefore, the approach of determining the internal state by calculation from voltage and current data was chosen rather than that of disassembling lithium-ion rechargeable batteries to measure their internal state. This choice was also intended to increase the applicability of the approach by reducing the training cost. In this way, it becomes possible to reduce the size of the extra margin necessary in design as well as to enhance convenience.

1. Introduction

Because of the high energy density of lithium-ion rechargeable batteries (LiB), there has been an increased need in recent years for these batteries as the power source for electric vehicles in the move toward a decarbonized society⁽¹⁾⁻⁽³⁾. Long-term durability performance of 10 years or more is sought for on-board LiB⁽¹⁾. The LiB performance degradation transition over the long term sometimes displays non-linear behavior that diverges from its linear behavior with respect to the power of the time period and number of charge-discharge cycles. Accurately predicting the prospects for that durability is no simple matter (Fig. 1)^{(4),(5)}. As to why the LiB performance degradation transition is non-linear, the reasoning here is that it may be because the multiple factors that cause expression of LiB performance differ one from the other in their degradation

behavior with respect to their elapsed time^{(4),(6)}. Generally, LiB performance degradation is predicted using methods that extrapolate it empirically using acquired data, or methods (acceleration tests) that shorten the time taken to

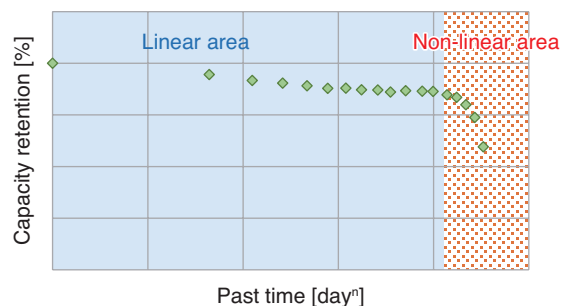


Fig. 1 Degradation transition into non-linearity

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confirm degradation behavior according to the intensity of degradation factors such as temperature and state of charge (SOC)^{(7),(8)}. In the former case, making long-term predictions with empirical models that have been trained using training data from a brief training period risks diminishing the reliability of the prediction results^{(3),(4),(9)}. Also, in the latter case, the balance of degradation between multiple factors of LiB performance expression changes, so that a simple acceleration of aging time is not achieved, and the degradation outlook cannot be readily judged, as has been previously suggested⁽¹⁰⁾. In order to address these risks, development would require a long development period and a large margin for durability design.

Meanwhile, research sought to address those issues by taking up physical models based on physical phenomena⁽¹¹⁾. These have the following two issues, however, so they are not purely physical models⁽⁹⁾. The first issue is that model training requires values for the physical properties involved, such as electrode plate thickness and electrolyte salinity. For that reason, users who did not design and manufacture the LiB cannot readily acquire those items of information, and creation of a model is no easy matter. The second issue is that because the principle formula is for an ideal state, it often does not match actual behavior. For this reason, the model formula uses correction factors that adjust for those differences, and the various behaviors are expressed by similar figures.

For the present research, therefore, a solution to these issues was sought by proposing a semi-physical model⁽⁴⁾ that makes the model portion that calculates LiB performance based on the factors in LiB performance expression into a physical model and makes degradation behavior in those various expressions into an empirical model that is trained from the test results. In this paper, Chapter 2 below will discuss a degradation simulation model that makes it possible to represent non-linear degradation behavior; Chapter 3 will describe that model's training method and accuracy verification results; and Chapter 4 will describe discussion using model technology in acceleration tests by means of heating.

2. Creation of a Degradation Simulation Model

2.1. Degradation Model Aims and Concept

The method of creating the semi-physical model will be described. First, the performance expression mechanism portion that is not dependent on the materials used in the LiB or on its design values is treated as a physical model portion. Next, the factor portion that is dependent on materials used and design values is treated as the empirical model portion that is trained mathematically on the basis of test results. For the empirical model training, the behavior of change with respect to the loads that are causes of degradation is measured for each of those factors by durability tests and used as training data for training

the model. By empirical model training in the degradation behavior for each factor, those portions that do not fit with a theoretical, ideal state can be made correctable, and since it does not require values for the various physical properties that are necessary to the physical model, its applicability can be heightened by comparison with the physical model. In addition, when attention is focused on each LiB performance expression factor, those factors are found to be single mechanisms and they display linear degradation transitions according to their respective theories. For example, the growth behavior of the solid electrolyte interphase (SEI) membrane that forms on the surface of the anode material and the electrolyte interface follows the square root law⁽¹²⁾. Consequently, predictions in regions that extend into longer times than the training data can be estimated with only slight deviation by straight-line extrapolation from the square root of the time. By these approaches, it becomes possible to represent non-linear degradation behavior with respect to time and charge-discharge cycles. Since LiB performance is expressed by a combination of multiple factors, cases where a single factor dominates degradation behavior indicate linear degradation. However, when that dominant factor switches over to another factor, then the degradation behavior is non-linear. For this reason, non-linear degradation behavior can be represented if performance expression factors are modeled individually and performance is calculated from the combination of factors based on their mechanisms. Since degradation is estimated virtually by calculation, it also becomes possible to remove the time constraint in investigations as compared to tests.

Changes in the LiB performance expression factors that become explanatory variables in the model are generally measured by disassembly analysis. With this kind of method, further durability tests of a sample cannot be conducted beyond the point when it has been disassembled. Consequently, training for one degradation transition behavior necessitates causing degradation of multiple samples under the same conditions, and fabricating samples in which degradation advances differently. For this method, acquiring training data necessitates many samples and many analyses. For the present investigation, therefore, an approach by analysis with current and voltage data that can be acquired without disassembling a battery was implemented.

2.2. Simulation Model Structure

2.2.1. Rated capacity model

In this clause, the configuration of a rated capacity model based on the expression mechanism for capacity will be explained. Figure 2 shows a conceptual diagram of the LiB rated capacity expression mechanism and calculation. As shown in the figure, expression of the rated capacity involves the relationship of the following four explanatory variables. By following this relationship, the rated capacity can be calculated⁽¹³⁾.

- (A) Cathode Ah-Open Circuit Potential (Ah-OCP) curve
- (B) Anode Ah-OCP curve
- (C) $OCV_{\max}$ [V]
- (D) $OCV_{\min}$ [V]

In other words, degradation of the rated capacity can be defined as occurring by means of changes in these variables (A)-(D). For this investigation, training in the change behavior in these variables (A)-(D) is carried out empirically. However, (C) can be defined as a voltage that is determined by the specifications, so here it was handled as a constant. The variable (D) is the open circuit voltage (OCV) for the current that is determined by the specifications that occurs when the LiB is discharged with a constant current up to the voltage similarly determined in the specifications, and its value changes under aging conditions in conjunction with the degradation of LiB internal resistance. For this investigation, therefore, $OCV_{\min}$ was taken as a function of the LiB internal resistance that accompanies degradation. The $OCV_{\min(t)}$ model formula isolates the resistance component ($R_{IMP(t)}$) of the frequency where the change from the initial value ($OCV_{\min(t_0)}$) has the highest correlation with the impedance (IMP) obtained from the cell and models it in Eq. (1)⁽¹³⁾. $R_{IMP(t)}$ in Eq. (1) is given according to the calculation results from the internal resistance model, which will be described later. In the equation, d is a correction factor that is learned in order to make $R_{IMP(t)}$ agree with the measured $OCV_{\min(t)}$ in Eq. (1).

$$OCV_{\min(t)} = OCV_{\min(t_0)} + d \times R_{IMP(t)} \quad (1)$$

For this investigation, the Ah axis of Ah-OCP is represented as the standard 0 Ah at the initial time of 0% SOC, which is represented as the basis Ah. From this it is possible to represent the rated capacity by the relationship in Eq. (2) below.

$$\text{Rated capacity}_{(t)} = f(\text{Cathode Ah-OCP curve}_{(t)}, \text{Anode Ah-OCP curve}_{(t)}, OCV_{\max}, OCV_{\min(t)}) \quad (2)$$

For the rated capacity_(t) at time t , the LiB characteristic

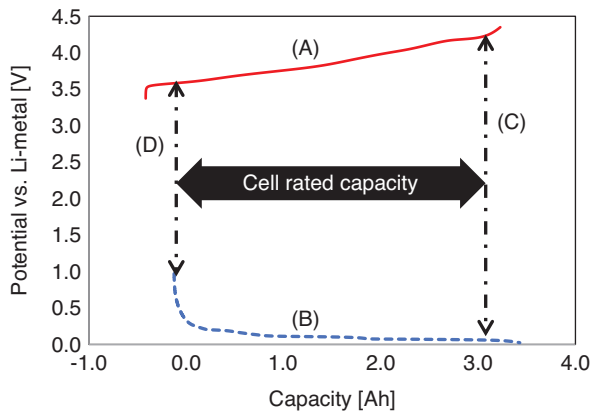


Fig. 2 Mechanism of capacity calculation

of Ah-OCV is determined from the combination of the electrical characteristic of the cathode (cathode Ah-OCP curve_(t)) and of the anode (anode Ah-OCP curve_(t)) at that same time. In addition, the Ah range that can be used as the LiB rated capacity is calculated from the OCV ($OCV_{\max}$) at 100% SOC and the OCV ($OCV_{\min(t)}$) at 0% SOC calculated from Eq. (1).

The change in variable (A) that accompanies degradation is represented by changing the basis Ah axis by multiplying that initial (A) by the capacity retention of the cathode. When changing the Ah-OCP curve, the respective electric potential positions of the cathode and anode according to the cell voltage during aging are fixed in place in the basis Ah axis direction, and the Ah-OCP curve profile is changed in the basis Ah axis direction (Fig. 3). Variable (B) also has changes made to it, as done with (A), by the anode capacity retention, in addition to which the anode Ah-OCP curve shifts to the higher-potential side with respect to the cathode Ah-OCP curve (the anode Ah shift) because of a side reaction of SEI formation, and variable (B) after degradation was represented by a parallel shift of the anode Ah-OCP curve with respect to the basis Ah axis⁽⁴⁾ (Fig. 4).

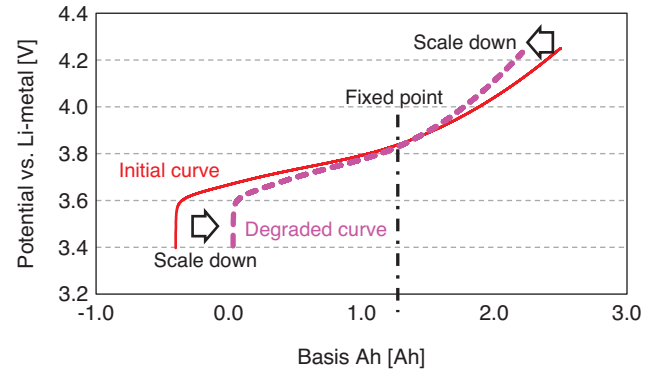


Fig. 3 Calculation of aging cathode curve

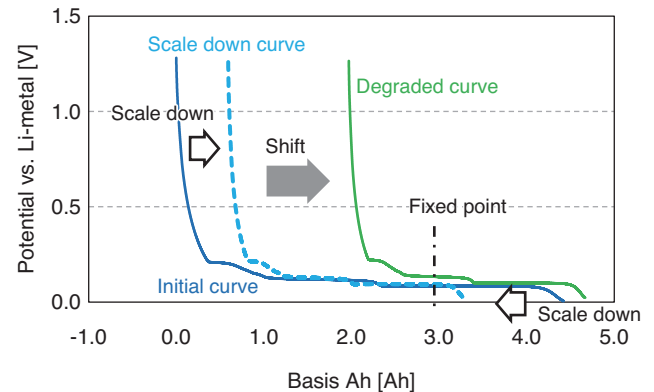


Fig. 4 Calculation of aging anode curve

2.2.2. Internal resistance model

In this clause, the configuration of a rated capacity model based on the expression mechanism for internal resistance will be explained. Figure 5 shows a conceptual diagram of the LiB internal resistance expression mechanism by means of a schematic cross-section of a cell. The LiB internal resistance is expressed by a combination of multiple components, and when power is supplied, internal resistance is generated from electron transfer (i)(viii), lithium ion insertion-extraction reaction (ii)(vii), and lithium ion diffusion migration (iii)-(vi) respectively, in that order.

For this investigation, LiB internal resistance is represented by the equivalent circuit shown in Fig. 6, and the following six items were treated as explanatory variables⁽¹⁴⁾.

- (R_0) Electron conduction resistance and ion conduction resistance [Ω]
- (R_1) Resistance component of anode reaction resistance [Ω]
- (CPE_1) Electric double-layer component of anode reaction resistance
- (R_2) Resistance component of cathode reaction resistance [Ω]
- (CPE_2) Electric double-layer component of cathode reaction resistance
- (CPE_3) Diffusion resistance

The sum of the resistances expressed by these six components is the internal resistance of the LiB. Its degradation occurs with the change of these components, similar to what is described in Clause 2.2.1. The six

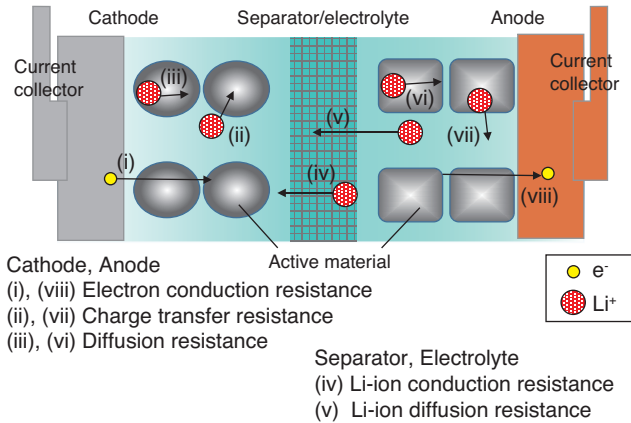


Fig. 5 Mechanism of internal resistance calculation

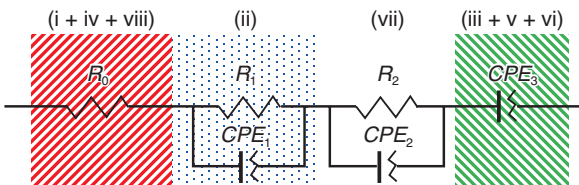


Fig. 6 Equivalent circuit of internal resistance

resistances are dependent on how long power is supplied, and since they can be represented by functions for the time power is supplied using the equivalent circuits, the calculation of the internal resistance of the LiB as a whole from their combination is performed as a physical calculation. On the other hand, the internal resistances that are treated as explanatory variables are trained empirically in change behavior acquired through tests. Having a modeled structure for each internal resistance component makes it possible to represent the changes in time dependency of reaction resistance and diffusion resistance in the cathode and anode accompanying degradation, and this has made it possible to estimate internal resistance with greater accuracy.

Internal resistance is generally evaluated by direct current resistance (DCR) that is calculated from the slope of the current-voltage (I-V) characteristic. For this model, therefore, it is output as a quantity converted to DCR from the equivalent circuit in Fig. 6. Due to the circumstances of the method for evaluating the I-V characteristic, it measures not only a pure resistance value, but also the change in OCV that accompanies the supply of power. For this model, therefore, the change in OCV that accompanies the change in SOC when power is supplied was also included, in addition to the overvoltage, in calculating the closed circuit voltage (CCV) that is generated by the application of current. By using the calculation results described above in Clause 2.2.1., the change in OCV (ΔOCV) that accompanies the change in the charge amount due to the supply of power was calculated, the I-V characteristic was calculated from the sum (ΔV_{total}) with overvoltage (ΔV) due to internal resistance, and in this way the accurate DCR was calculated (Fig. 7). In addition, the overvoltage (ΔV) due to internal resistance was calculated [Eq. (3)] by solving it as a function of time (t) from the equivalent circuits in Fig. 6⁽¹³⁾.

$$CCV = OCV - I \times \left[R_0 + R_1 \times \left\{ 1 - \exp \left(\frac{-t}{R_1 \times C_1} \right) \right\} + R_2 \times \left\{ 1 - \exp \left(\frac{-t}{R_2 \times C_2} \right) \right\} + \frac{t^{CPE_3}}{CPE_{3-T} \times CPE_{3-p}!} \right] + \Delta OCV \quad (3)$$

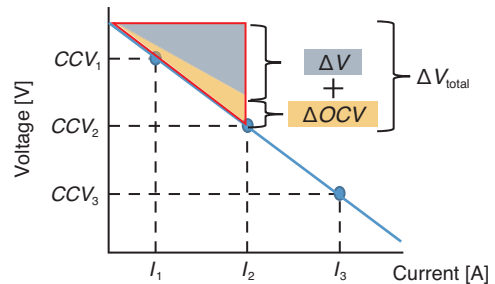


Fig. 7 How to calculate I-V characteristics

CPE has the T value that represents the electric double-layer capacity and the p value that represents the state of collapse of arcs in the Nyquist plot. Therefore, those values in CPE_n ($n = 1 - 3$) are indicated in the equation by CPE_{n-T} and CPE_{n-p} .

When calculating the transient voltage response that accompanies the application of current, the calculation cannot be done using the parallel circuits with CPE and R used in the equivalent circuits in Fig. 6. Therefore, Eq. (4) was used to convert CPE_n and the resistance component R_n connected in parallel with it into C_n and perform the transient response calculation.

$$C_n = CPE_{n-T} \frac{1}{CPE_{n-p}} \times R_n \frac{1 - CPE_{n-p}}{CPE_{n-p}} \quad (4)$$

3. Model Training and Simulation Results

3.1. Training Method

In this chapter, an overview will be given of the training method used for the empirical model portion. For this investigation, a single-factor experimental method based on experimental design was applied to conduct durability tests by multiple standards of each factor that influences degradation and acquire training data⁽¹⁵⁾. The durability tests were conducted with load conditions other than degradation factors corresponding to explanatory variables taken as constants. The LiB degradation factors assumed for the present investigation are shown in Table 1.

The method of analysis by non-disassembly used to isolate the explanatory variables for each model will be described later in the respective clauses. The degradation behavior of each explanatory variable acquired was fitted to below candidate equations Eqs. (5) and (6) and the better fit was selected.

$$y = a \times x^b + c \quad (5)$$

$$y = a \times \ln(x) + c \quad (6)$$

For this, a was derived for each durability condition and b for conditions of resting degradation and of degradation of the supply of power, deriving them as a constant with a single common value for each explanatory variable respectively. Next, the derived coefficient a was a curved surface fitted on a multidimensional surface that has the number of degradation factors as the number of dimensions and a map of degradation coefficients was created. For this investigation, durability tests for the purpose of model training had a load applied for a durability period of four months and the two months necessary for measuring performance changes occurring during that time, making a total of six months. Performance measurements were made at the start of aging and then during aging in the first week, the second week, the fourth week, and at four-week intervals thereafter, a total of eight times.

Table 1 List of degradation factors

Factor		Degradation factor
Rated capacity	Cathode capacity	Temperature, Average cathode SOC, Cathode SOC width, Cathode current density (Charge, Discharge)
	Anode capacity, Ah shift	Temperature, Average anode SOC, Anode SOC width, Anode current density (Charge, Discharge)
Internal resistance	R_0	Temperature, Average cell voltage, Cell voltage width, Cell current density (Charge, Discharge)
	R_1, CPE_1	Temperature, Average anode SOC, Anode SOC width, Anode current density (Charge, Discharge)
	R_2, CPE_2	Temperature, Average cathode SOC, Cathode SOC width, Cathode current density (Charge, Discharge)
	CPE_3	Temperature, Average cell voltage, Cell voltage width, Cell current density (Charge, Discharge)

3.1.1. Method of analysis of explanatory variables in rated capacity model

This clause will describe the method of analysis of each explanatory variable used in the rated capacity model⁽¹³⁾. In order to acquire the explanatory variables (A)-(D) described above in Clause 2.2.1. without disassembling a battery, dV/dQ analysis and charge-discharge curve analysis were used. The anode capacity was isolated (Fig. 8) by preparing a cell of the same specifications in its initial state in advance, removing the anode plate from it, obtaining the Ah-dV/dQ curve from that anode plate, and fitting that curve so that its peaks matched the positions of the peaks originating in the anode (arrow symbols in the figure) in the Ah-dV/dQ curve obtained from the cell.

Next, the curve of the cathode potential in the cell is calculated by subtracting the anode Ah-closed-circuit potential (CCP) curve defined in Fig. 8 from the charging curve obtained from the cell. In order to match this curve, the Ah-CCP curve of the cathode, obtained in advance as was done with the anode, is fitted with the curve, and in this way the cathode capacity was isolated. Using the cathode and anode Ah-CCP curves determined above, the Ah shift amount was isolated from the change in the Ah position in the same potential before and after aging.

3.1.2. Method of analysis of explanatory variables in internal resistance model

This clause will describe the method of analysis⁽¹⁴⁾ of each explanatory variable used in the internal resistance model.

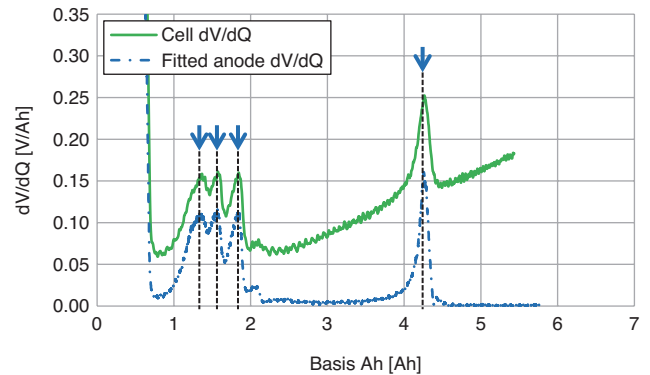


Fig. 8 Anode fitting

Generally, electrochemical impedance spectroscopy (EIS) is used as the method for separating LiB internal resistance components. With on-board LiB, however, the internal resistance values are extremely small, at several milliohms, and the expression frequencies for cathode and anode reaction resistance at normal temperatures are extremely close together. For this reason, the respective cathode and anode reaction resistances cannot readily be derived from IMP data obtained at normal temperatures. In addition, output demand is large, the current rate that flows during output is large, and the time is long. This means that the diffusion resistance component, for which the IMP is obtained in a static state by EIS, cannot be accurately acquired by that method as a dynamic value during output. For this investigation, therefore, conduction resistance (R_0) was isolated by IMP analysis using EIS at normal temperature, the individual reaction resistances (R_1 , CPE_1 , R_2 , and CPE_2) were isolated by low-temperature IMP analysis⁽¹⁶⁾, and diffusion resistance (CPE_3) was isolated by analysis of transient voltage response data acquired during I-V characteristic evaluation at normal temperature matched with the characteristics of each resistance component. The equivalent circuits shown in Fig. 6 were used to identify each resistance component. For CPE_3 , values to match the transient response for voltage with respect to applied current were derived by the least-squares method using an equivalent circuit configured with R_0 , R_1 , CPE_1 , R_2 , and CPE_2 values identified by each type of IMP analysis.

3.1.3. Simulation method

Figure 9 shows the simulation flow. The voltage and current that are the load conditions are converted from the LiB internal state at that time (t) into the loads on the cathode and anode, respectively, and provided as input

Table 2 Model formula and differential equation

	Model formula	Differential equation	
n-th power type	$y = a \times x^b + c$	$\frac{dy}{dx} = \frac{a^{1/b} \times b}{(y - 1)^{(1/b - 1)}}$	
Natural log type	$y = a \times \ln(x) + c$	$\frac{dy}{dx} = a \times e^{-(1-y/a)}$	Time model : $x = \text{time}$ Energization model: $x = \text{Ah}$

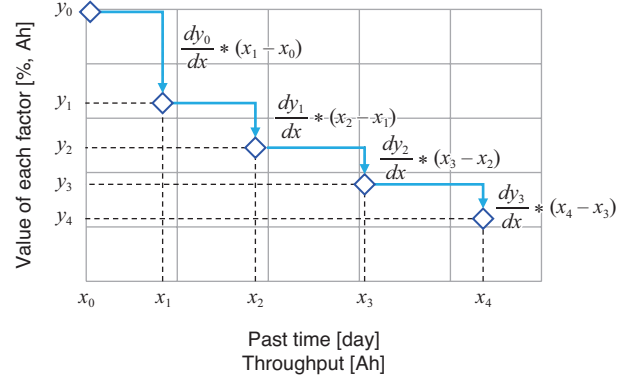


Fig. 10 Factor amount calculation method

for each model formula. The internal state is also updated with the results calculated in each model formula and the calculation for the next time ($t+1$) is carried out. The differential equation (Table 2) for calculating the amount of change per unit period that can be defined according to the model formula determined for each explanatory variable is used for each model, the coefficient a determined by the load conditions is read into it to calculate the amount of change per unit time, and those results are integrated to calculate the degradation behavior (Fig. 10)⁽¹⁵⁾.

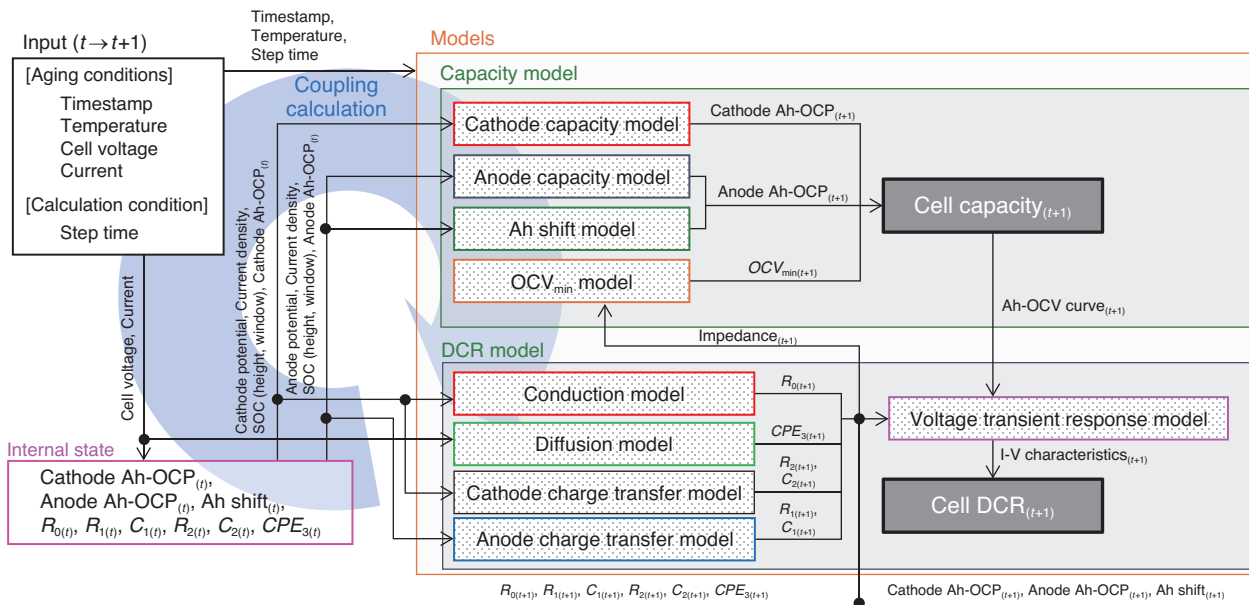


Fig. 9 Simulation flow

For degradation due to the passage of time, Miner's rule was used during the step period where degradation is added up in the simulation to calculate the degradation coefficient and basic SOC by time-weighted average. For the degradation due to supply of power, the degradation coefficient and basic SOC were calculated according to the respective pulse loads that were isolated, and the amount of change per unit section to be calculated was added up⁽¹³⁾.

3.2. Simulation Results

3.2.1. Capacity model

Figure 11 shows the results for calculation of changes in degradation during durability tests. Estimation of degradation by an empirical model (for this investigation, a power law model is used to conduct comparative evaluation) diverges from the measurement results as aging progresses, but this model achieved calculation results in line with measurement results even in the region at the end of aging where degradation is shown to be non-linear.

3.2.2. Internal resistance model

As in Clause 3.2.1., the results for calculation of changes in degradation are shown in Fig. 12. As with the capacity

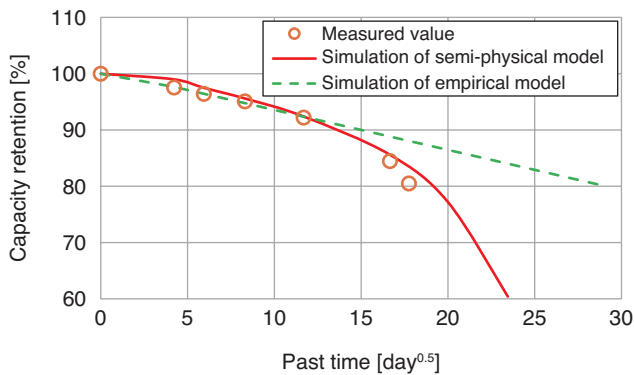


Fig. 11 Verification result of capacity model

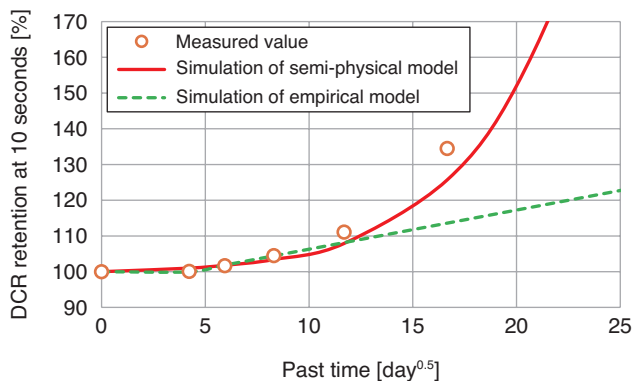


Fig. 12 Verification result of DCR model

model, this model achieved calculations in line with measurement results even at the end of aging, in contrast with the empirical model.

3.3. Conclusion

Verification results demonstrate that this simulation model has made it possible to represent non-linear behavior in the changes in degradation over the long term, which was not possible with the conventional empirical model. In addition, the condition raising the load of aging conditions successfully showed that the pattern of changes in degradation and the state of degradation inside the LiB are different.

4. Representation of Internal Degradation State under Concentration Condition

4.1. Capacity Model

Figure 13 shows a comparison of degradation behavior estimated with the model by changing only the temperature load. This was done in order to confirm differences in the state of internal degradation by acceleration tests. When changes in degradation are compared, degradation behavior under high-temperature conditions shows non-linear behavior at the time (arrow symbol in the figure) in the vicinity of 85% capacity retention where a bend occurs. At low temperature, by contrast, the bend timing does not occur and linear behavior is shown. This suggests that the same degradation pattern does not occur in acceleration testing by heating. Table 3 shows the degradation state of each factor at 60% capacity retention under both conditions. It is apparent from this that even when capacity retention is the same, the amount of degradation is different for each factor, and in the two tests with different temperatures, the degradation states are not the same. These investigation results indicate that in acceleration tests that raised the aging load, the internal degradation state and different behaviors of the patterns of changes in degradation accompanying it are possible to represent with this simulation technology.

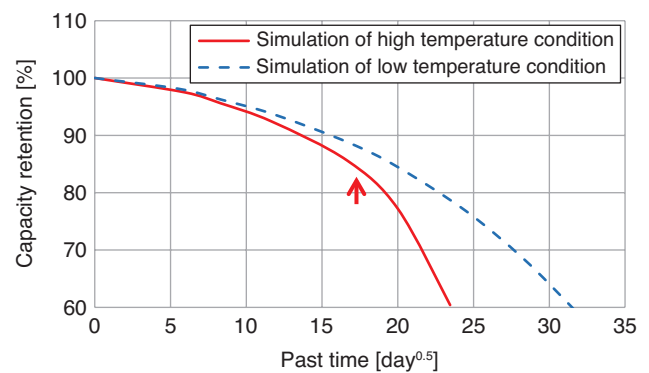


Fig. 13 Difference in capacity degradation behavior due to acceleration test

Table 3 Simulation result at capacity retention of 60%

	Aging condition	
	High temperature	Low temperature
Cathode capacity retention	85.2%	87.0%
Anode capacity retention	31.7%	59.6%
Ah shift value	0.11 Ah	0.09 Ah

Table 4 Simulation result at DCR retention of 150%

	Aging condition	
	High temperature	Low temperature
Ohmic resistance retention	100.0%	100.0%
Cathode's reaction resistance retention	111.0%	117.7%
Anode's reaction resistance retention	211.0%	179.9%
Diffusion resistance retention	181.9%	273.7%

4.2. Internal Resistance Model

As with the capacity model, the results for estimation of differences in degradation behavior in acceleration tests are shown in Fig. 14 and Table 4. In internal resistance degradation behavior, a bend occurred in the changes in degradation under both conditions, and the behavior under different timing of that occurrence (DCR retention value) was calculated (arrow symbols in Fig. 14). Table 4 shows the degradation state of each factor at the point when the DCR retention is 150% under both conditions. It is apparent from this, as with the capacity model, that even when the DCR retention is the same, the degradation state is not the same in the two tests at different temperatures.

These results indicate that even in internal resistance, there are differences in degradation behavior (Fig. 14) and the internal state after degradation (Table 4) under acceleration conditions, and that it is possible to represent those differences with this simulation technology.

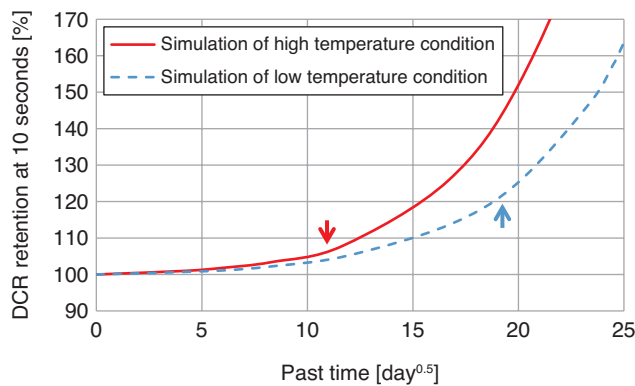


Fig. 14 Difference in capacity degradation behavior due to acceleration test

5. Summary

A semi-physical model was created that has a physical structure based on the performance expression mechanism and an empirical structure that uses data measured in tests for training. This made possible prediction of long-term degradation behavior that is highly accurate by comparison with the power law model, which is the general empirical model. In addition, this approach is capable of representing changes in degradation and grasping internal degradation states that could not be readily realized by acceleration tests with increased load, thereby suggesting that this is a method that can be used to evaluate durability performance. By the use of this technology, it becomes possible to predict the prospects for durability with accuracy in new LiB development. In addition, where up to now major constraints had been imposed on the range of usable voltage, current, and temperature in order to secure an extra margin for durability design, this technology was able to reduce these constraints, yielding the expectation of heightened convenience in terms of an expanded cruising range and an enhanced acceleration performance for electric vehicles. Moreover, this technology is applicable not only to use in development, but also to the used car market and the reuse market, where it can be applied to the diagnosis of residual value from different perspectives, not only for estimating the remaining life expectancy of a LiB over the long term, but also for matching to applications by battery life prediction that takes the internal degradation state into consideration, and so on. This technology can be expected to serve as a tool that will be able to support the widespread adoption of electric vehicles and LiB reuse.

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