Lifetime Prediction for Heavy-duty Industrial Lithium-ion Batteries that Enables Highly Reliable System Design

Makoto Abe
Katsunori Nishimura, Dr. Sc.
Eiji Seki
Hiroshi Haruna
Tokiyoshi Hirasawa
Shingo Ito
Tadashi Yoshiura

OVERVIEW: The operating life of the batteries is a major factor in the reliability and cost of energy storage systems such as those used as backup power supplies or for the reduction of generated power fluctuations from renewable energy sources. This article describes research aimed at improving the accuracy of lifetime prediction for lithium-ion batteries by devising a lifetime prediction technique able to handle three variables simultaneously (number of cycles, operating time, and ambient temperature). The work included formulating a new lifetime prediction equation that combines the square root function law, Arrhenius law, and additive law approaches used in the past. The benefits of this prediction technique include both reducing system cost by minimizing the number of batteries required, and improving reliability by providing a clearer understanding of the margin for error. Use of this technique to predict the operating life of batteries in energy storage systems contributes to reduced costs, improved reliability, and other benefits.

INTRODUCTION

THE high energy density and compact and lightweight construction of lithium-ion batteries ("batteries") make them candidates for use in energy storage systems such as those used as backup power supplies or for the reduction of generated power fluctuations from renewable energy power generation systems. Being able to reliably predict the lifetimes of the batteries used in these applications makes energy storage systems more affordable and gives greater confidence in the estimated operating life of the systems. However, previous techniques were unable to consider three relevant variables for predicting lifetimes simultaneously, namely, the number of cycles, operating time, and ambient temperature, and this made them inadequate for providing accurate predictions of battery lifetime.

In response, Hitachi has formulated a prediction equation that incorporates all three of these variables, and has developed a technique able to predict battery lifetimes accurately.

This article describes a lifetime prediction technique for heavy-duty industrial lithium-ion batteries that allows highly reliable system design.

BATTERY LIFETIME AND PREVIOUS PREDICTION TECHNIQUE

Repeated charging and discharging of batteries causes a gradual decay in their capacity (see Fig. 1). When designing energy storage systems, it is necessary to take this decay into account when selecting the initial capacities to ensure that the batteries continue to satisfy the required design capacity throughout their operating lives. To achieve this, it is important to have a technique available that can accurately predict battery lifetime (the point in the battery's life when its capacity falls to the level of the design capacity). In turn, accurate prediction of lifetime requires an understanding of the processes

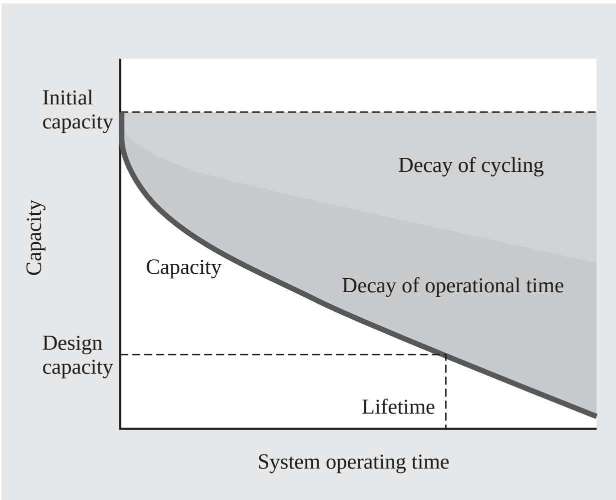

Fig. 1—Battery Lifetime Definition and Capacity Degradation.
Repeated charging and discharging of batteries causes their capacity to deteriorate. The rates of progress of the two different forms of degradation, "decay of cycling" and "decay of operational time," are considered independent of each other.

that cause capacity to decay, and the formulation of a prediction equation that uses appropriate variables.

It is understood that two processes in particular account for the decay in battery capacity. The first is the degradation caused by expansion and contraction of the anode and cathode materials during charging and discharging (called "decay of cycling"), which progresses with each charge/discharge cycle. The second is degradation due to chemical reactions inside the battery (called "decay of operational time"), which progresses over time regardless of whether the battery is charged and discharged. It is believed that these two types of degradation are largely independent of each other, and that the overall decay in battery capacity is the sum of the two effects. This means that the battery's operating time needs to be considered as well as the number of cycles.

As it is known that both "decay of cycling" and "decay of operational time" proceed more rapidly at higher temperatures, the ambient temperature also needs to be taken into account.

Therefore, accurate prediction of battery lifetime requires a prediction equation based on three variables: number of cycles, operating time, and ambient temperature.

The following describes the three main prediction techniques used in the past.

Prediction based on the square root function law estimates the contribution of "decay of cycling" to the battery lifetime by extrapolating capacity based on the assumption that the decay in battery capacity is proportional to the square root of the number of cycles[(1)]. The square root function law method can also estimate the contribution of "decay of operational time" to the battery lifetime by assuming that the decay in battery capacity is proportional to the square root of the operating time.

Prediction of battery lifetime based on the Arrhenius law uses a square root function law to approximate the battery's capacity decay at different temperatures and assumes that the coefficient of the square root function law at each temperature is proportional to an exponential function of temperature[(2)].

Prediction of battery lifetime based on an additive law adds together the contributions of the "decay of operational time" and "decay of cycling" obtained using the square root function laws[(3)].

Prediction using a square root function law on its own can only deal with one or other of "decay of cycling" and "decay of operational time," and is unable to take account of the increase in each type of deterioration caused by higher temperature. Similarly, the Arrhenius law and additive law methods are also unable to deal with all three variables (number of cycles, operating time, and ambient temperature) at the same time. This makes these past techniques inadequate for the accurate prediction of battery lifetime. In response, in the work described in this article, Hitachi developed a technique for the accurate prediction of battery lifetime by combining these previous techniques to formulate a new lifetime prediction equation that uses all three variables.

DEVELOPMENT OF LIFETIME PREDICTION TECHNIQUE

Formulation of Lifetime Prediction Equation

The new lifetime prediction equation combines the concepts from the previous techniques (see Fig. 2). The equation represents the "decay of operational time" as a function of the square root of operating time, and the "decay of cycling" as a function of the square root of the number of cycles. The ambient temperature T is one of the factors that determines the rate at which the "decay of operational time" and "decay of cycling" occur, and is represented in the equation as an Arrhenius (exponential) function. The equation calculates the values for "decay of operational time" and "decay of cycling" separately and then subtracts their sum from 1 to express the battery capacity as a capacity retention ratio (proportion of the battery's initial 100% capacity).

The variables in the equation are the number of cycles (N_{cyc}), operating time (N_p), and temperature (T), and the result is the capacity retention ratio (Q_R). The constants in the equation are the gas constant (R) and four constants (A_p, A_{cyc}, E_p, and E_{cyc}) that characterize the rates for "decay of operational time" and "decay of cycling." These latter four constants are determined by battery testing, consisting of storage tests to determine the constants for "decay of operational time" and cycle tests to determine the constants for "decay of cycling," each of which is conducted for three different ambient temperatures (25°C, 35°C, and 45°C).

$$Q_R = 1 - \left(\underbrace{A_P \cdot e^{-\frac{E_P}{RT}} \cdot \sqrt{N_P}}_{\text{Decay of operational time}} + \underbrace{A_{cyc} \cdot e^{-\frac{E_{cyc}}{RT}} \cdot \sqrt{N_{cyc}}}_{\text{Decay of cycling}} \right)$$

Fig. 2—New Lifetime Prediction Equation.
The equation calculates the capacity retention ratio (Q_R) from the number of cycles (N_{cyc}), operating time (N_p), and temperature (T).

To obtain the *Ap* and *Ep* constants for the "decay of operational time," the results of the storage tests at each temperature were approximated by a square root function law and plotted on an Arrhenius plot with the logarithm of the square root function law coefficient as the vertical axis and the inverse of temperature as the horizontal axis.

The results of the cycle tests were analyzed in the same way to obtain the constants for "decay of cycling." However, because the time taken to perform a cycle test means that the results also include a "decay of operational time" component in addition to the "decay of cycling," the results were adjusted to include the "decay of cycling" component only when calculating the constants. To obtain this component, the time required for the cycle test was substituted into the equation for "decay of operational time" described above (see Fig. 2) to calculate the "decay of operational time," and this value was then subtracted from the cycle test results. Using the "decay of cycling" component obtained from this calculation, the "decay of cycling" constants (A_{cyc} and E_{cyc}) were then obtained using the same procedure as for "decay of operational time."

Comparison of Lifetime Prediction Equation with Test Measurements

The suitability of the lifetime prediction equation was verified by comparing it against the results of a long-term cycle test. The long-term cycle test ran for two and a half years, during which the batteries experienced 6,000 cycles. Fig. 3 shows the actual measurement results from the long-term cycle test as white dots and the predictions as a gray line. The results show a close agreement between the measured and predicted trend in the capacity retention ratio, indicating that the newly developed lifetime prediction equation is suitable for use.

EXAMPLE APPLICATIONS OF LIFETIME PREDICTION

The new lifetime prediction technique can be used to reduce system costs because it allows the installed battery capacity to be minimized in systems that in the past would have required a large safety margin. Furthermore, an understanding of how much margin to allow for factors such as the number of cycles and ambient temperature provides benefits such as higher system reliability. The following sections describe some actual implementations of the technique.

Lifetime Prediction

Fig. 4 shows battery lifetime predictions for a constant ambient temperature (25°C) and number of cycles (four per day). Under these conditions, the predicted capacity retention ratio after 10 years was 75%.

This result can be used to determine the battery capacity required by an energy storage system. For example, if the design capacity of a system were

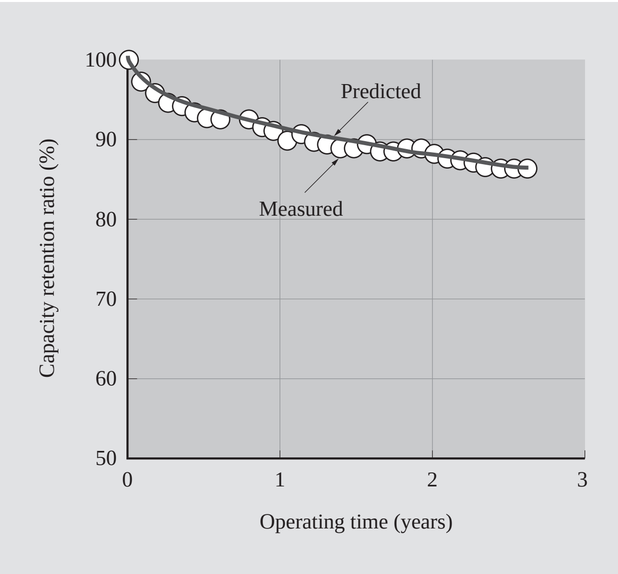

Fig. 3—Results of Battery Testing.
The graph shows close agreement between the predicted and actual results of a long-term cycle test.

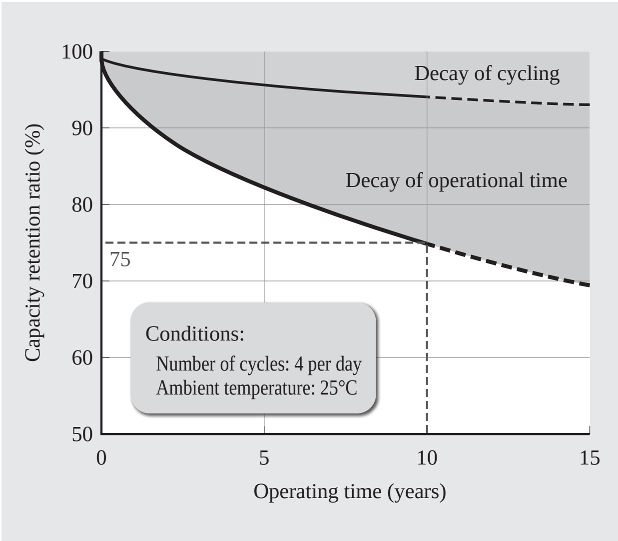

Fig. 4—Results of Lifetime Prediction.
The graph shows the predicted lifetime under standard conditions for ambient temperature and number of cycles. Accurate prediction of battery lifetime reduces the system cost.

chosen to correspond to a capacity retention ratio of 75%, ensuring that this level of minimum capacity is maintained over a period of 10 years would require an initial capacity equal to 1.33 times the design capacity (1.33 being the inverse of 75%).

As indicated by this example, an accurate prediction of battery lifetime reduces the system cost by allowing the installed capacity to be minimized, instead of requiring a large safety margin as was the case in the past.

The lifetime prediction equation also allows the relative proportions of the "decay of operational time" and "decay of cycling" to be compared. In the example shown in Fig. 4, the comparison indicates that the "decay of operational time" predominates over the "decay of cycling."

Margin Allowance

Fig. 5 shows comparisons of the prediction results for cases when either the ambient temperature or the number of cycles was held constant and the other parameter varied.

The study found that varying the number of cycles from four to eight per day at an ambient temperature of 25°C had little effect on the behavior of the capacity retention ratio. On the other hand, when the batteries were subjected to four cycles per day, the capacity retention ratio after 10 years fell to 75% for an ambient temperature of 25°C, but to 60% for an ambient temperature of 32°C.

As this example indicates, an understanding of how much margin to allow for the number of cycles and ambient temperature helps enhance system reliability because it allows the system to be designed in a way that maintains the design capacity even when these parameters vary from day to day.

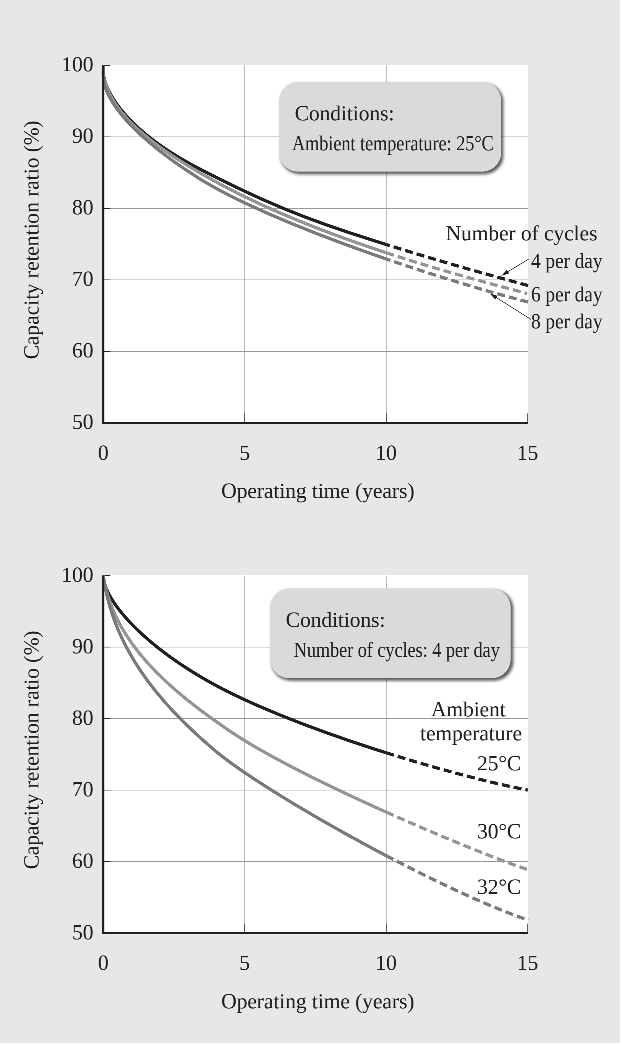

Fig. 5—Margin Allowance.
The graphs show the results for cases when either the ambient temperature or the number of cycles was held constant and the other parameter varied. An understanding of how much margin to allow for the number of cycles and ambient temperature helps enhance system reliability.

CONCLUSIONS

This article has described a lifetime prediction technique for heavy-duty industrial lithium-ion batteries that allows highly reliable system design.

Hitachi has developed a lifetime prediction technique that takes account of three variables at once, namely, number of cycles, operating time, and ambient temperature. Use of this new technique reduces the system cost by allowing the installed battery capacity to be minimized, instead of requiring a large safety margin as was the case in the past. Furthermore, an understanding of how much margin to allow for factors such as the number of cycles and ambient temperature provides benefits such as higher system reliability. In the future, Hitachi intends to use this technique to predict battery lifetimes in energy storage systems to help make them more affordable and reliable.

While ambient temperature is one of the variables considered by the prediction, heat generated in the battery itself means that the ambient temperature and battery temperature are not necessarily the same. Therefore, it is also necessary to consider the amount of generated heat, which is dependent on operating conditions. Also, it is not possible to take account of factors such as causes of capacity degradation that occur unpredictably in battery operation. It will be necessary in the future to expand the scope of lifetime prediction by developing prediction equations that take account of these factors.

ACKNOWLEDGMENTS

The research described in this article was part of a project for Development of an Electric Energy Storage System for Grid-connection with New Energy Resources in which Hitachi participated under a contract from the New Energy and Industrial Technology Development Organization (NEDO). The authors would like to take this opportunity to express their thanks.

REFERENCES

(1) T. Yoshida et al., "Degradation Mechanism and Life Prediction of Lithium-Ion Batteries," Journal of The Electrochemical Society **153 (3)**, A576-A582 (2006).

(2) H. Yoshida et al., "Capacity Loss Mechanism of Space Lithium-Ion Cells and its Life Estimation Method," Electrochemistry **71 (12)**, 1018 (2003).

(3) Y. Kobayashi et al., "Development of Performance Analysis Technique for Lithium-ion Batteries Used to Smooth Interconnection with Power Grid," 51st Battery Symposium in Japan Abstracts, p. 117, The Committee of Battery Technology, The Electrochemical Society of Japan (Nov. 2010) in Japanese.

ABOUT THE AUTHORS

Makoto Abe

Joined Hitachi, Ltd. in 1992, and now works at the Department of Battery Research, Materials Research Center, Hitachi Research Laboratory. He is currently engaged in the research and development of battery electrolytes.

Katsunori Nishimura, Dr. Sc.

Joined Hitachi, Ltd. in 1992, and now works at the Department of Battery Research, Materials Research Center, Hitachi Research Laboratory. He is currently engaged in the research and development of battery electrolytes. Dr. Nishimura is a member of The Electrochemical Society of Japan (ECSJ).

Eiji Seki

Joined Hitachi, Ltd. in 1994, and now works at the Department of Battery Research, Materials Research Center, Hitachi Research Laboratory. He is currently engaged in the research and development of battery anode materials.

Hiroshi Haruna

Joined Hitachi, Ltd. in 2003, and now works at the Department of Battery Research, Materials Research Center, Hitachi Research Laboratory. He is currently engaged in the research and development of battery electrolytes. Mr. Haruna is a member of The Chemical Society of Japan.

Tokiyoshi Hirasawa

Joined Shin-Kobe Electric Machinery Co., Ltd. in 1978, and now works at the Tsukuba Research Laboratory, Hitachi Chemical Co., Ltd. He is currently engaged in the research and development of lithium-ion batteries for industry. Mr. Hirasawa is a member of the ECSJ and The Institute of Electrical Engineers of Japan.

Shingo Ito

Joined Shin-Kobe Electric Machinery Co., Ltd. in 2007, and now works at the Energy Devices and Materials Development Center, Tsukuba Research Laboratory, Hitachi Chemical Co., Ltd. He is currently engaged in the research and development of lithium-ion batteries for industry.

Tadashi Yoshiura

Joined Shin-Kobe Electric Machinery Co., Ltd. in 2008, and now works at the Energy Devices and Materials Development Center, Tsukuba Research Laboratory, Hitachi Chemical Co., Ltd. He is currently engaged in the research and development of lithium-ion batteries for industry.