



## Statistical methodology for predicting the life of lithium-ion cells via accelerated degradation testing

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### ABSTRACT

Statistical models based on data from accelerated aging experiments are used to predict cell life. In this article, we discuss a methodology for estimating the mean cell life with uncertainty bounds that uses both a degradation model (reflecting average cell performance) and an error model (reflecting the measured cell-to-cell variability in performance). Specific forms for the degradation and error models are presented and illustrated with experimental data that were acquired from calendar-life testing of high-power lithium-ion cells as part of the U.S. Department of Energy's (DOEs) Advanced Technology Development program. Monte Carlo simulations, based on the developed models, are used to assess lack-of-fit and develop uncertainty limits for the average cell life. In addition, we discuss the issue of assessing the applicability of degradation models (based on data acquired from cells aged under static conditions) to the degradation of cells aged under more realistic dynamic conditions (e.g., varying temperature).

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### 1. Introduction

Accurate life prediction is a crucial component to the successful commercialization of high-power battery technologies for various applications, including the automotive industry. In conjunction with the Partnership for a New Generation of Vehicles (PNGV), the Advanced Technology Development (ATD) Program was initiated in 1998<sup>1</sup> by the U.S. Department of Energy (DOEs) Office of Advanced Automotive Technologies to find solutions to the barriers that limit the commercialization of high-power lithium-ion batteries for hybrid-electric vehicle (HEV) and plug-in hybrid (PHEV) applications. A significant barrier that limits commercialization is the requirement for a long-lived battery (15 years or more). Thus, one goal of the ATD Program is to develop methods to accurately predict the life of lithium-ion batteries in the HEV and PHEV environments with a high level of confidence given only 1 or 2 years of accelerated aging. As part of this effort, a number of methodologies for estimating life (with associated uncertainty bounds) have been developed and applied to degradation data that were

acquired during various accelerated degradation experiments. The purpose of this article is to describe our current methodology for estimating cell life with uncertainty bounds. This methodology, which depends on degradation and error models, is illustrated by application to data acquired during a recent calendar-life experiment. In this particular case, the forms of the degradation and error models were simple and empirically based. The general approach outlined here should be applicable to other model forms as well. Monte Carlo simulations were used to assess lack-of-fit and to indicate the uncertainty limits for average cell life (assuming that no lack-of-fit is found). Finally, we discuss the issue of whether or not models based on data acquired from cells aged under static conditions will accurately predict cell degradation in real-life, dynamic aging conditions. An experimental process for exploring this issue is discussed.

### 2. Development of degradation and error models

As the cell ages, the performance degrades. The level of performance degradation depends on the time and stresses to which the cell has been exposed. The measured performance at any given time for an individual cell is, thus, a combination of effects that can be related to the technology, to the unique behavior of the individual cell, and to the measurement process. A generic model form that captures these effects is given by  $Y_i(X;t) = \mu(X;t) + \gamma_i(X;t)$ ,

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where  $Y_i(X;t)$  represents the measured performance of the  $i$ th cell after being subjected to aging for time  $t$  under stress factors that are represented by  $X$ . The average cell performance is represented by  $\mu(X;t)$ . The combined effects that are related to the unique behavior of the  $i$ th individual cell and measurement error are represented by  $\gamma_i(X;t)$ . Separate model forms for  $\mu(X;t)$  and  $\gamma_i(X;t)$  are presented here.

A degradation model expresses the *expected* performance level versus the time and conditions under which a cell has been aged:  $\mu(X;t)$ . This model can be empirical, chemistry/physics-based, or some combination of both. A wide variety of model forms are possible. In practice, the specific form of the model that is used will depend on the particular technology and set of stress factors. In the calendar-life example given in this paper we use a simple degradation model with a single stress factor, temperature. The simple model and its log transform are given in Eqs. (1) and (2), respectively. Here,  $\mu(X;t) = \mu(T;t)$  represents the expected relative resistance (resistance at time  $t$  divided by the initial resistance).

$$\mu(T;t) = 1 + \exp \left\{ \beta_0 + \beta_1 \cdot \frac{1}{T} \right\} \cdot t^\rho \quad (1)$$

$$\log(\mu(T;t) - 1) = \beta_0 + \beta_1 \cdot \frac{1}{T} + \rho \cdot \log(t) \quad (2)$$

Temperature is represented by  $T$  while  $\beta_0$ ,  $\beta_1$ , and  $\rho$  represent the model parameters. This particular model form has both a physical and empirical basis. The physical basis is apparent through the Arrhenius ( $1/T$ ) rate dependence.

We used the convention that  $\mu(X;t) = 1$  for  $t = 0$  and then increases in value as the cell ages. Note that, in cases where the natural response is initially one and decreases to zero as the cell ages,  $\mu(X;t)$  can be considered as a model for the inverse of the natural response. Examples of such a natural response include relative power and relative capacity. Thus in such cases,  $\mu(X;t)$  can be considered as a model for inverse relative power and inverse relative capacity.

In order to conduct accurate simulations, one needs a viable error model that accounts for variability due to measurement error as well as the intrinsic differences between cells. We recommend the error model given in Eq. (3).

$$\gamma_i(X;t) = \delta_i \cdot (\mu(X;t) - 1) + \pi_i(t) \quad (3)$$

where  $\delta_i$  represents a random, cell-specific, proportional effect and  $\pi_i(t)$  represents the effects of measurement error on  $Y_i(X;t)$ . The unique effect of the  $i$ th cell with respect to performance (e.g., relative resistance) is given by the product,  $\delta_i \cdot (\mu(X;t) - 1)$ . This effect increases as the  $i$ th cell ages since  $(\mu(X;t) - 1)$  increases with  $t$ . Considered over the population of cells, this effect models the increasing level of cell-to-cell variation in performance that is observed as cells age under the same stress condition. The overall difference between the observed and expected performance ( $\gamma_i(X;t)$ ) for the  $i$ th cell is a combination of the cell-effect ( $\delta_i \cdot (\mu(X;t) - 1)$ ) and the effects of measurement error ( $\pi_i(t)$ ). We regard measurement error to be associated with the measurement system and to be independent of the state of the cell. When  $Y_i(X;t)$  represents relative resistance,

$$Y_i(X;t) = \frac{R_{\text{true}}(i,t) + \varepsilon_i(t)}{R_{\text{true}}(i,0) + \varepsilon_i(0)} \quad (4)$$

where  $R_{\text{true}}(i,t)$  is the unknown, but true, value of the resistance of the  $i$ th cell at time  $t$ , and  $\varepsilon_i(t)$  is the specific unknown error associated with that measurement. Thus,

$$\pi_i(t) = \frac{R_{\text{true}}(i,t) + \varepsilon_i(t)}{R_{\text{true}}(i,0) + \varepsilon_i(0)} - \frac{R_{\text{true}}(i,t)}{R_{\text{true}}(i,0)} \quad (5)$$

**Table 1**  
Experimental conditions

Temperature (°C)	State-of-charge	Number of cells
30	SOC <sub>MAX</sub>	3
40	SOC <sub>MAX</sub> - 10%	3
40	SOC <sub>MAX</sub>	3
40	SOC <sub>MAX</sub> + 10%	3
47.5	SOC <sub>MAX</sub> - 10%	3
47.5	SOC <sub>MAX</sub>	3
47.5	SOC <sub>MAX</sub> + 10%	3
55	SOC <sub>MAX</sub> - 10%	3
55	SOC <sub>MAX</sub>	3
55	SOC <sub>MAX</sub> + 10%	3

If we assume that (1) the measurement errors are independent with a relative standard deviation of  $\alpha$  (i.e.,  $\sigma_\varepsilon = \alpha \cdot R_{\text{true}}(i,0)$ ); (2)  $R_{\text{true}}(i,t)/R_{\text{true}}(i,0) \approx 1$ ; and (3)  $\alpha$  is relatively small, then the variance of  $\pi_i(t)$ , given by  $\sigma_\pi^2$ , is approximately  $2 \cdot \alpha^2$ . In practice, we expect  $R_{\text{true}}(i,t)/R_{\text{true}}(i,0) < 1.5$  and  $\alpha < 0.05$ . From here on, the variance of  $\delta_i$  will be denoted by  $\sigma_\delta^2$ . Assuming that the mean values of  $\varepsilon_i(t)$  and  $\delta_i$  are zero, then within a given group of cells that have experienced the same stresses and aging time,

$$\text{Mean}(Y_i(X;t)) = \mu(X;t) \quad (6)$$

and

$$\text{Var}(Y_i(X;t)) = \text{Var}(\gamma_i(X;t)) = \sigma_\delta^2 \cdot (\mu(X;t) - 1)^2 + \sigma_\pi^2 \quad (7)$$

Thus, this error model implies that the expected variability of the cells increases as the expected level of degradation increases. Here,  $\mu(X;t)$  refers to the particular degradation model that is used, not necessarily that from the example. In the case of the example, the degradation and error models were used with experimental data to estimate the model parameters,  $\beta_0$ ,  $\beta_1$ ,  $\rho$ ,  $\sigma_\pi^2$  and  $\sigma_\delta^2$ .

The error model provides a basis for assessing the level of measurement error based on the experimental aging data. There are other more direct ways to assess the level of measurement error (e.g., see [1]). Idaho National Laboratory (INL) has developed a direct approach to determining the level of measurement uncertainty, and has taken significant steps towards reducing the level of that uncertainty [2,3]. Prior to cell aging, the test equipment is calibrated according to manufacturer specifications and independently checked for accuracy at different current and voltage levels within the full-scale range of the test channel using a digital voltmeter and shunt (to determine current). The test channel response measured by the digital voltmeter and shunt, and their associated calibration errors, are used to determine the total channel error (in terms of voltage and current). The uncertainties of these measured responses are then used to assess the level of measurement uncertainty of any derived performance variable (e.g., cell impedance) required by the life model. Specifically, the uncertainties of the derived variable(s) are based on low-order Taylor Series approximations of the derived variable(s) with respect to the independent measurements of voltage and/or current. In the example, we compare the levels of measurement uncertainty obtained from the error model and from the direct approach given in [2,3].

### 3. Experimental

The purpose of the experiment was to validate the testing recommended in the Technology Life Verification Test (TLVT) Manual [4]. High-power SAFT VL7P lithium-ion cylindrical cells were purchased for this experiment. These cells were rated at 7 Ah with a maximum and minimum voltage of 4.0–2.7 V, respectively. Ten experimental conditions were investigated with a total of 30 cells (see Table 1). Two controlled factors (aging temperature and state

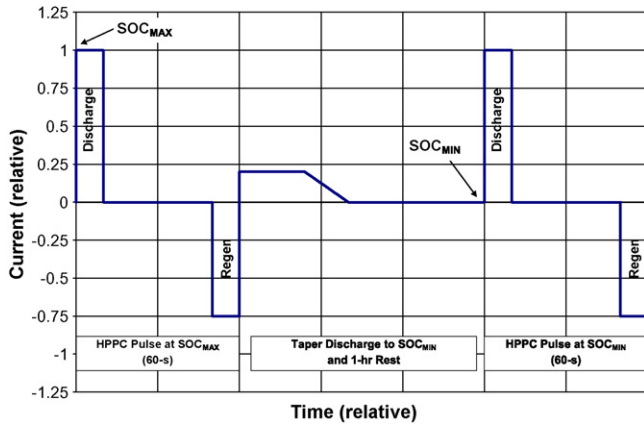


Fig. 1. MPPC pulse profile.

of charge) were used. Nine experimental conditions were derived from a full-factorial experimental plan involving three levels for temperature (40 °C, 47.5 °C, and 55 °C) and three levels for state of charge ( $SOC_{MAX} - 10\%$ ,  $SOC_{MAX}$ , and  $SOC_{MAX} + 10\%$ ) as defined below. The values for  $SOC_{MAX}$  and  $SOC_{MIN}$  for the SAFT VL7P cells were voltage based, and roughly corresponded to 62% and 24% SOC, respectively. Additionally, three cells were evaluated at 30 °C and  $SOC_{MAX}$  for the purpose of validation.

The cells were subjected to calendar-life testing at the designated temperatures with a daily taper charge back to the voltage corresponding to the designated SOC. The cells were placed in an isothermal chamber during aging to ensure temperature control. Calendar-life testing was interrupted every 31.5 days for reference performance testing (RPT) at 30 °C to gauge the performance degradation of the cell. Eight-hour temperature soaks followed each RPT. The RPT consisted primarily of the minimum pulse power characterization (MPPC) test [4]. The pulse profile is shown in Fig. 1, and consists of 10 s discharge and regen (i.e., charge captured during regenerative braking) pulse at specified rates (typically a 5C<sub>1</sub> discharge rate) with a 40 s rest in between at two different SOC conditions ( $SOC_{MAX}$  and  $SOC_{MIN}$ ). These conditions are usually specified by the manufacturer and are intended to represent the anticipated operating range of the battery during normal usage. The purpose of this test was to acquire useful resistance data while avoiding effects that may artificially age the cells more rapidly such as time spent at full charge. Although this profile was shown to have deficiencies [5], the data are still useful for the purposes of developing a life and error model and demonstrating the methodology to predict life. From the MPPC test, discharge (and regen) resistances can be calculated by taking the difference between the initial voltage prior to the pulse and the voltage at the end of the pulse divided by the constant current. In summary, data discussed in this article represent the discharge resistances at  $SOC_{MAX}$  through approximately 221 days of calendar-life aging with seven RPTs.

## 4. Results

### 4.1. Fitted degradation model

The data from the full-factorial part of the design were used to construct a degradation model. The 30 °C data were used strictly for model validation. The effect of SOC (over its experimental range) on degradation was not found to be statistically significant. Thus, the degradation model described by Eq. (2) was fit to the experimental data (40 °C, 47.5 °C, and 55 °C) using robust linear regression [6]. In cases where the degradation model is not expressed as a linear

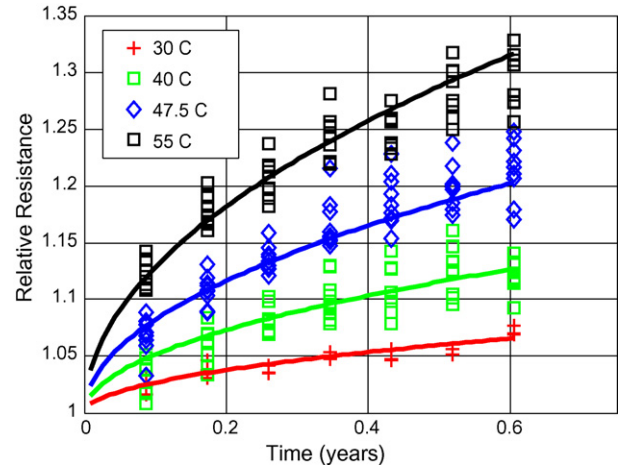


Fig. 2. Fitted degradation model with data.

model, parameter estimation can be more difficult. Nevertheless, there are a number of methods that exist for estimating parameters in the case of nonlinear models (e.g., see [7]).

The final fitted degradation model is  $\hat{\mu}(X; t) = 1 + \exp\{\hat{\beta}_0 + \hat{\beta}_1 \cdot (1/T) \cdot t^{1/2}\}$ , where  $\hat{\beta}_0 = 18.11(1.1)$  and  $\hat{\beta}_1 = -6236(360)$ ; the “bootstrap” standard error of each estimated parameter is given in parentheses (see Section 4.4). Fitting the full model,  $\mu(X; t) = 1 + \exp\{\beta_0 + \beta_1 \cdot 1/T\} \cdot t^\rho$ , resulted in  $\hat{\rho} = 0.52$  with a standard error of 0.019. Since  $\hat{\rho}$  was indistinguishable from 1/2,  $\rho$  was fixed at 1/2 in order to simplify the model with a common transformation of time. The experimental data (symbols) and final fitted degradation model (lines) are given in Fig. 2. Note that the 30 °C data are well-approximated by the model.

The model was used to estimate the lifetime of the cell at a specified temperature by specifying an end-of-life criterion for the cell. Suppose that the lifetime ( $t_{EOL}$ ) is defined to be the time at which the resistance is predicted to increase by 30% when the temperature,  $T$ , is 303 °K, then  $\hat{t}_{EOL} = \exp\{(\log(0.3) - (\hat{\beta}_0 + \hat{\beta}_1 \cdot 1/T))/0.5\} = 12.6$  years. The “bootstrap” standard error of the estimated lifetime is 1.9 years (see Section 4.4). Note that the range of applicability of this model is limited to conditions supported by the experimental data. For example, it would not be appropriate to use this model to predict the time associated with a 100% resistance rise since the maximum resistance rise observed during the experiment was less than 35%. Thus, one should continue the degradation experiment until at least some of the test cells reach the end-of-life condition. Finally, note that  $\hat{t}_{EOL}$  is an estimate of the mean cell life of the technology that was tested. The lifetimes of individual cells will vary about  $t_{EOL}$ .

### 4.2. Fitted error model

Fig. 3 shows the results of fitting the error model described by Eq. (3) to summaries of the experimental data (40 °C, 47.5 °C, and 55 °C) using robust linear regression. Each symbol summarizes the sample variance of observed relative resistance versus the square of the difference between the expected relative resistance and unity for each group of data partitioned by temperature and RPT. The high level of scatter about the robust regression line (red) is consistent with the statistical variation of the sample variance based on sparse data. The estimated error model parameters are given by the slope and half of the intercept of the fitted line in Fig. 3; they are  $\hat{\sigma}_\delta^2 = 3.2 \times 10^{-3}$  and  $\hat{\alpha}^2 = 1.2 \times 10^{-4}$ , respectively. Thus, the cell-specific proportional effect is estimated to have a standard deviation of about 0.06 and the measurement error is estimated

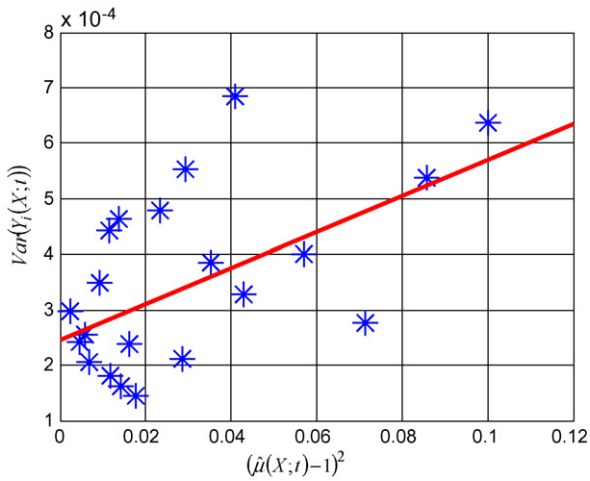


Fig. 3. Fitted error model. Data represented are across all SOC levels.

to have a relative standard deviation of about 0.01. As a point of comparison, the independent determination of measurement error using the INL methodology found that the relative standard deviation of the measurement error (depending on the test equipment) was about 0.006.

4.3. Lack-of-fit statistic

Lack-of-fit with respect to the fitted degradation model was measured using the statistic,  $SS_{LOF} = \sum_{i=1}^3 \sum_{t=1}^7 n_{it} / \hat{\sigma}_{it}^2 \cdot (\bar{Y}_{it} - \hat{\mu}_{it})^2$ , given 3 treatment groups (temperatures) and 7 RPTs. Here,  $\bar{Y}_{it}$  is the average relative resistance of the  $i$ th temperature group at time  $t$  (consisting of  $n_{it}$  cells);  $\hat{\mu}_{it} = 1 + \exp(\hat{\beta}_0 + \hat{\beta}_1 \cdot 1/T_i) \cdot t^{1/2}$ ; and  $\hat{\sigma}_{it}^2 = \hat{\sigma}_\delta^2 \cdot (\hat{\mu}_{it} - 1)^2 + \hat{\sigma}_\pi^2$ . Monte Carlo simulations (see Section 4.4) based on the developed degradation and error models were used to assess whether the computed value of the lack-of-fit statistic (in this case,  $SS_{LOF} = 52.0$ ) is unusually large under the assumption that the models are accurate.

Fig. 4 summarizes the distribution of the values of  $SS_{LOF}$  obtained from 1000 Monte Carlo simulation trials of the experiment with the assumed models (see Section 4.4). It is interesting to compare  $SS_{LOF}$  with the simulated distribution of  $SS_{LOF}$  values. The empirical cumulative distribution function evaluated at  $SS_{LOF} = 52.0$  exceeds 0.99. This means that less than 1% of the values of  $SS_{LOF}$  from the

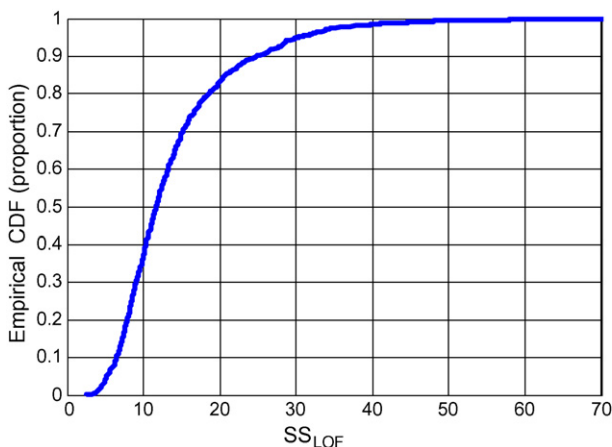


Fig. 4. Empirical cumulative distribution function of  $SS_{LOF}$  from simulations.

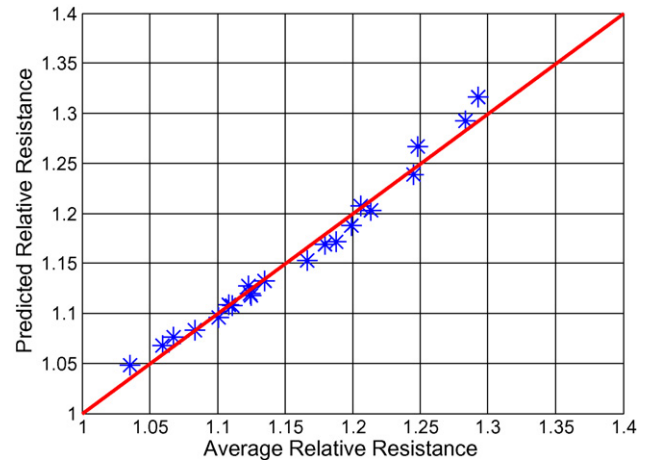


Fig. 5. Predicted relative resistance versus average relative resistance.

simulation exceeded 52.0. Thus, it is unlikely that one would obtain such a large value of  $SS_{LOF}$  by chance given that the models are accurate. So, we conclude that there is significant statistical evidence for lack-of-fit.

The nature of the lack-of-fit reflected in the large value of  $SS_{LOF}$  is illustrated in Fig. 5. The model seems to slightly over-predict relative resistance at high levels of degradation, perhaps providing a conservative estimate of life. In accepting this model and life estimates derived from it, one needs to carefully consider this evidence for lack-of-fit from a practical perspective.

4.4. Monte Carlo simulations

Monte Carlo simulations are used to simulate the measured performance of cells subjected to aging at various stress conditions. Accurate degradation and error models are required in order to accurately simulate performance. The simulations can be used to assess the uncertainty of model parameters and functions of model parameters (e.g., mean cell life) estimated from experimental data. In addition, the simulations can provide a basis for assessing the quality of the model based on “lack-of-fit” statistics.

The simulation (see Appendix A for details) is set up to mimic the actual experiment: same test duration, RPT frequency, experimental conditions, and number of cells per condition. A large number of independent simulation trials are performed in which different random realizations of cell-to-cell effects and measurement errors (assumed to be normally distributed) are added to the assumed truth provided by the degradation model. Thus, each trial simulates the actual experiment that was performed. For each trial, model parameters and average cell life are re-estimated. In addition, the “lack-of-fit” statistic,  $SS_{LOF}$ , is re-computed. Across all trials, distributions of re-estimated model parameters, re-estimated average cell life, and re-computed  $SS_{LOF}$  are obtained. Based on the distributions of re-estimated model parameters and re-estimated average cell life, one can assess the statistical uncertainty of model parameters and cell life estimated from fitting the actual experimental data. In addition, the value of  $SS_{LOF}$  obtained from fitting the original data can be compared with the distribution of re-computed  $SS_{LOF}$  across trials to assess lack-of-fit.

Fig. 6 shows the simulated data and fitted model (evaluated at 40, 47.5, and 55 °C) associated with a single simulation trial. The degradation model parameters re-estimated based on the simulation data from this single trial are  $\hat{\beta}_0 = 16.82$  and  $\hat{\beta}_1 = -5820$ . Corresponding estimates of the error model parameters for this trial

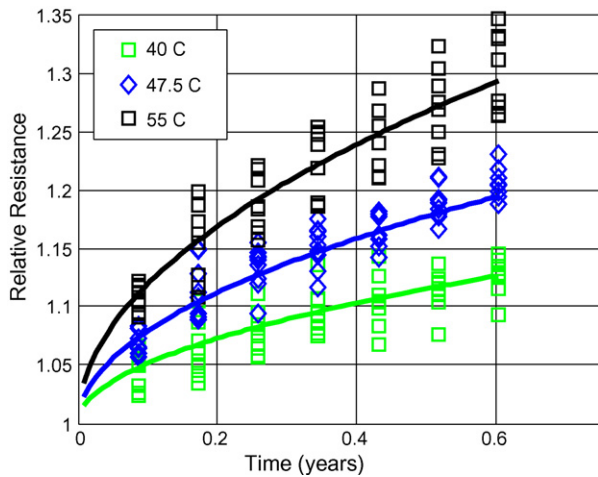


Fig. 6. Simulated data and fitted degradation model.

are  $\hat{\sigma}_\theta^2 = 7.8 \times 10^{-3}$  and  $\hat{\sigma}^2 = 8.7 \times 10^{-5}$ . The corresponding mean cell lifetime is re-estimated to be  $\hat{t}_{EOL} = 10.7$  years with  $SS_{LOF} = 8.7$ .

Fig. 7 summarizes the distribution of estimated mean cell lifetime for the complete set of 1000 simulation trials from which the “bootstrap” standard error [8] of the estimated lifetime was derived. This figure provides an idea of the level of statistical uncertainty associated with estimating cell life given such an experiment and assuming that the forms of the degradation and error models are accurate. A lower 90% bootstrap confidence limit for mean cell lifetime is given by the 10th percentile of the 1000 simulated estimates of life which is approximately 10.3 years. Thus, assuming the forms of the degradation and error models are accurate, we have 90% confidence that the actual mean cell life is at least 10.3 years. This can be compared with the life estimate based on the actual experimental data which is 12.6 years.

### 5. Degradation in dynamic aging conditions

As is usually the case, the calendar-life model presented here was developed with data collected from cells aged under static stress conditions. It is recommended that some degree of additional testing be conducted in order to assess whether or not the models can accurately predict cell degradation in real-life, dynamic aging conditions (e.g., varying temperature). This issue is discussed in Refs. [9] and [10]. For

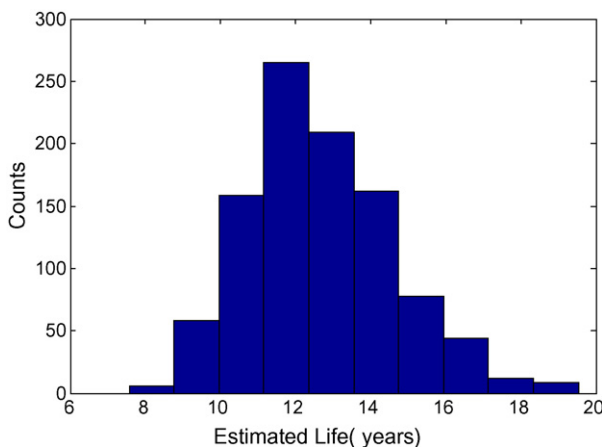


Fig. 7. Histogram of life estimates (1000 simulation trials).

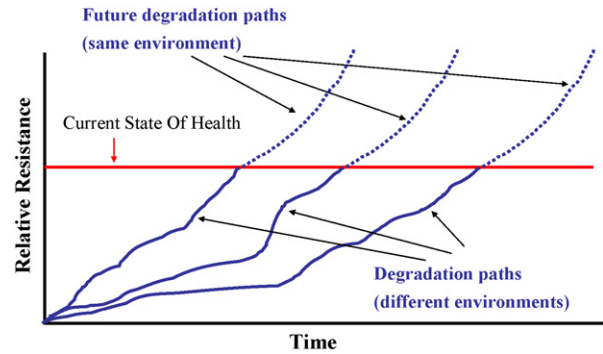


Fig. 8. Illustration of memoryless process.

example, our degradation model can be solved in terms of  $t$ , resulting in  $t^{-1/2} = \exp\{\beta_0 + \beta_1 \cdot 1/T\}/(\mu(X; t) - 1)$ . Furthermore,  $\partial\mu(X; t)/\partial t = 1/2 \cdot \exp\{\beta_0 + \beta_1 \cdot 1/T\} \cdot t^{-1/2} = \exp\{2 \cdot (\beta_0 + \beta_1 \cdot 1/T)\}/[2 \cdot (\mu(X; t) - 1)]$ . By integrating this derivative equation and allowing temperature to vary over time, we have  $\int_1^\mu (m - 1)dm = 1/2 \int_0^t \exp\{2 \cdot (\beta_0 + \beta_1 \cdot 1/T(\tau))\}d\tau$  which simplifies to  $1/2\mu^2 - \mu + 1/2 = 1/2 \int_0^t \exp\{2 \cdot (\beta_0 + \beta_1 \cdot 1/T(\tau))\}d\tau$ . Thus, if a cell was subjected to a temperature profile given by  $T(\tau)$ , then the expected relative resistance would be the solution to  $\hat{\mu}$ , where  $1/2\hat{\mu}^2 - \hat{\mu} + 1/2 = 1/2 \int_0^t \exp\{2 \cdot (\hat{\beta}_0 + \hat{\beta}_1 \cdot 1/T(\tau))\}d\tau$ .

By using such an approach, a necessary condition for accurate prediction of cell degradation in dynamic conditions is that degradation be a “memoryless” process in terms of the performance measure that is used. A memoryless process is one in which the rate of future degradation does not depend on the environment that produced the current state of health, but instead, depends only on the current state of health and the future environment. The notion of a memoryless process that is discussed here has a clear connection to (and was motivated by) the idea of a Markov process in the context of probability theory (e.g., see [11]). Fig. 8 illustrates a memoryless process in which three cells reach a common state of health (relative resistance) at different times after being exposed to different environments. Once reaching the common level of relative resistance, the cells are subjected to the same environment for some additional amount of time. If the future degradation of the cells does depend on the past history, then “relative resistance” (by itself) is not a sufficient indicator of “state of health.” If so, the degradation models may provide inaccurate predictions of cell degradation in dynamic conditions. Furthermore, other performance measures (possibly in combination) will be required to evaluate the “state of health” of a cell.

There are a number of simple experiments that might be performed to assess whether or not cell degradation as measured by relative resistance (or some other measure of performance) is a memoryless process. As indicated in Fig. 8, one could subject cells to a number of different environments. By using frequent measurements, each cell could be removed from testing and stored cold at the point when some common target level of performance is reached. Then, degradation of all cells would resume and be observed under a common environment. Significant differences in the subsequent degradation across the cells would indicate that the degradation process is not memoryless. We expect to conduct experiments like this in the future.

### 6. Conclusions

This paper provides methodology for modeling the degradation of lithium-ion cells and estimating mean cell life. The methodology

requires both a degradation model and an error model. Specific degradation and error models are discussed in the context of an illustrative example. In addition, this paper provides and illustrates methodologies (involving Monte Carlo simulations) for assessing lack-of-fit and assessing the statistical uncertainty associated with the predicted life.

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### Appendix A. Simulation details

The general simulation approach (which is a variant of the parametric bootstrap procedure [8]) is illustrated by expanding the generic modeling notation to  $Y_{ij}(t) = \mu(X_j; t) + \delta_{ij} \cdot (\mu(X_j; t) - 1) + \pi_{ij}(t)$ , where  $j$  represents the stress condition and  $ij$  represents the  $i$ th cell within the  $j$ th stress condition. Here,  $Y_{ij}(t)$  represents the measured relative resistance of the  $ij$ th cell at time  $t$ ,  $\mu(X_j; t)$  represents the expected relative resistance for cells under the  $j$ th stress condition at time  $t$ ,  $\delta_{ij}$  represents the random proportional effect of the  $ij$ th cell, and  $\pi_{ij}(t)$  represents the effect of the random measurement errors on relative resistance associated with the  $ij$ th cell at the initial measurement and at time  $t$ . The last term can be notionally partitioned into two terms:  $\pi_{ij}(t) \sim \lambda_{ij}(0) + \lambda_{ij}(t)$ , where the two terms represents the effect of the individual measurement errors on relative resistance. For these simulations, the random effects,  $\delta_{ij}$ ,  $\lambda_{ij}(0)$ , and  $\lambda_{ij}(t)$ , are assumed to be independent and normally distributed each with a mean of zero and variance of  $\sigma_\delta^2$ ,  $\alpha^2$ , and  $\alpha^2$ , respectively. In terms of the example described in Sections 3 and 4, the fitted degradation and error models are assumed to be true. That is,  $\mu_{it} = 1 + \exp\{\beta_0 + \beta_1 \cdot 1/T_i\} \cdot t^{1/2}$  and  $\sigma_{it}^2 = \sigma_\delta^2 \cdot (\mu_{it} - 1)^2 + 2 \cdot \alpha^2$ , where  $\beta_0 = 18.11$ ,  $\beta_1 = -6236$ ,  $\sigma_\delta^2 = 3.2 \times 10^{-3}$ , and  $\alpha^2 = 1.2 \times 10^{-4}$ .

The simulation is constructed as follows

1. Select  $J$  stress conditions to be used as well as number of cells per stress condition:  $\{n_j; j = 1:J\}$ .
2. Select the times at which the cells are to be measured:  $\{t_k; k = 1:K\}$ .
3. Compute the degradation model for each combination of stress condition and measurement time:  $\{\mu(X_j; t_k); (j = 1:J) \times (k = 1:K)\}$ .
  - For each trial, complete steps 4–8.
4. Obtain a random sample of proportional effects where the  $\delta_{ij}$  are sampled independently from a normal distribution with mean zero and standard deviation  $\sigma_\delta$ :  $\{\delta_{ij}; (i = 1:n_j) \text{ with } (j = 1:J)\}$ .
5. Obtain a random sample of initial measurement error effects where the  $\lambda_{ij}(0)$  are sampled independently from a normal distribution with mean zero and standard deviation  $\alpha$ :  $\{\lambda_{ij}(0); (i = 1:n_j) \text{ with } (j = 1:J)\}$ .
6. Obtain a random sample of measurement error effects where the  $\lambda_{ij}(t)$  are sampled independently from a normal distribution with mean zero and standard deviation  $\alpha$ :  $\{\lambda_{ij}(t_k); (i = 1:n_j) \text{ with } (j = 1:J) \text{ and } (k = 1:K)\}$ .
7. Combine the constituent effects from steps 3–6 to form the simulated relative resistance data:  $Y_{ij}(t_k) = \mu(X_j; t_k) + \delta_{ij} \cdot (\mu(X_j; t_k) - 1) + \lambda_{ij}(0) + \lambda_{ij}(t_k)$ . (a) Check that  $Y_{ij}(t_k) > 1$ .
8. Model the collective set of simulated resistance data for the current trial
  - Estimate model parameters (degradation and error).
  - Estimate average cell life.
  - Compute  $SS_{LOF}$ .
9. Compute summary statistics (e.g., standard deviations and order statistics) of model parameters, estimated cell life, and  $SS_{LOF}$  across trials. The standard deviations of the model parameters and estimated cell life are referred to as bootstrap standard errors.

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