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서로 다른 가속기법의 결합을 통한 2차 전지 사이클 시험 시간의 단축

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Cycle-life Test Time Reduction in Secondary Rechargeable Batteries by Combining Different Types of Acceleration

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신뢰성 평가 시험은 종종 성능 평가에 장기간의 시간이 요구되며, 전체 생산비용까지 증가시키는 문제점을 안고 있다. 스트레스를 이용한 가속수명시험은 제품의 신뢰성 고장과 밀접한 관련이 있는 고장 메커니즘의 촉진을 통해 고장에 이르는 기간을 단축함으로써 신뢰성 평가의 효율성을 도모할 수 있다. 본 연구에서는 이러한 스트레스 가속 시험에 빈도가속(Usage-Rate Acceleration) 또는 판정가속(Tightening Critical-Values) 등을 결합하여 한층 높은 가속효과를 도모하는 방법을 제안하고, 국내에서 생산되고 있는 2차 전지 제품에 대한 실제 시험 사례분석을 통해 결합된 가속방법의 효과를 실증적으로 보여주고 있다.

Keywords : Accelerated Life Test, Lithium Ion Battery, Cycle Life, Stress Acceleration, Tightened Critical-Values, Likelihood Ratio Test, Acceleration Factor

1. Introduction

In keeping pace with the era of multimedia, the electronic industries strive to be the first in developing and marketing various consumer-oriented information devices such as mini-computer, digital camcoder, PDA, and cellular phone, etc.

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The competitive development of merchandise has become possible through the small high-capacity secondary rechargeable battery technology, which can be considered as the core of multimedia products. Developed by Sony in 1991, lithium-ion batteries, which can be considered as a revolution in the battery industry, overcome several weaknesses, such as low capacity, heavy weight, and pollution, that traditional nickel cadmium (Ni-Cd) and nickel metal hybrid (Ni-MH) secondary batteries suffer from. As the name suggests, lithium ion batteries use lithium, the most lightweight metal on earth. Due to their demonstrated excellent energy density and cycle-life, lithium-ion batteries in a short time span have taken the largest part of the commercial markets for powering high-end electronics applications, including portable phones, camcorders, and computers [3].

On the other hand, developing a reliable product is the survival key in the keen competition for market occupancy among manufacturers. The reliability of a product is closely related to its life. It often takes a long time to assess the life of a product, causing major difficulties for manufacturers who, in turn, need to reduce product development time as much as possible. In order to reduce the test time, the accelerated life test method is generally used. There are three different types of acceleration in the literature: usage-rate acceleration, stress acceleration, and tightening critical-values. In these method, products are tested/or evaluated under severer conditions that are severer than the normal operating conditions to hasten product failure within a short time. Product life under the normal condition could then be estimated based on the result of the accelerated test. For a general discussion and detailed information on degradation models and analysis techniques, see [8, 9].

In general, the cycle-life of battery is generally the most important characteristic. It is closely related to the customers' CTQ (Critical To Quality). It usually takes quite a long time to complete the cycle-life tests for qualification. In order to shorten the testing time, battery engineers have struggled to devise various approaches, one of which has been a stress accelerated test using high temperature (e.g., [2, 11]). However, current market preferences are for batteries that perform well even at elevated temperatures (e.g., [4]). Further, multi-stresses accelerated tests such as temperature-voltage may often be not applicable for safety-related issues (e.g., fire and explosion). Consequently, the exploitation of a single type acceleration only may no longer be effective. One possible way to overcome such difficulties is to combine different types acceleration to expedite a product's failure during the test intervals. In other words, accelerating stresses, coupled with other types of acceleration, may lead to a shorter test time. Yang and Yang [14] and Yang [13], for example, considered the accelerated test situation where the tightened critical-value and the usage-rate acceleration were combined to further shorten the required test time, then they developed optimal plans for such tests.

In this study, we explore three types of acceleration in the cycle-life tests of the secondary rechargeable batteries: a new attempt to combine different types of acceleration to reduce the test time.

2. Technical Background

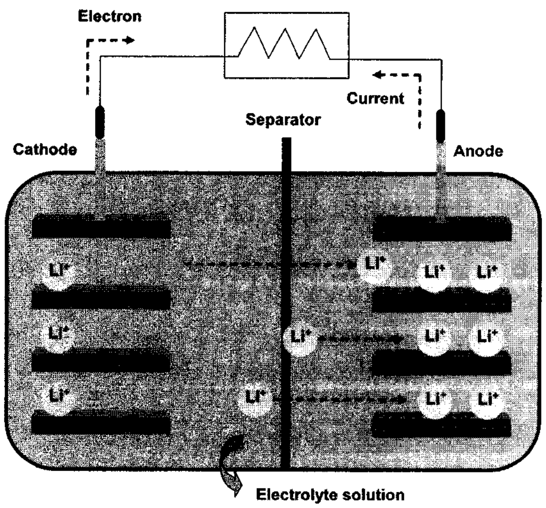
The test units in this application are samples of prismatic 950-mAh lithium ion batteries used for power device for mobile phones. The sample is shown in <Figure 1>.

<Figure 1> Test samples

A lithium-ion battery system is composed of four basic elements : the cathode (positive electrode, lithium compounds), anode (negative electrode, carbon), electrolyte solution, and separator. The basic structure of a typical lithium-ion battery system is shown in <Figure 2>. While the battery is being charged, lithium-ions moving from the cathode, leaving it with a net negative charge, are forced onto the anode, giving it a positive charge. During discharge, the ions flow in the opposite direction, from the anode to the cathode. The lithium ion battery uses the current generated during discharge. Because this reaction is reversible in the secondary battery but impossible in the primary battery, secondary batteries are capable of being recharged and reused up to hundreds of cycles (one cycle represents one charge/discharge).

The basic performance of a battery is characterized by its capacity, which is generally defined as the amount of charge available, expressed in ampere-hours (Ah). The cycle-life is defined as the number of complete charge/discharge cycles before the nominal capacity of the battery drops below the pre-specified value of its initial capacity. Although it is desirable that the battery retains the initial capacity as much as possible during usage time, in the batteries currently available, the capacity does tend to decrease through repetitive charge/discharge cycles. A number of efforts to evaluate and improve cycle-life of batteries have been taken [2-4, 7, 10].

As the cycle proceeds, the capacity of a test unit is repeatedly measured at every cycle through an automatic charge/discharge system, and its degradation is continuously monitored during the tests. In the cycle-life tests for qualification, each unit is subject to checked whether its capacity reaches a fixed threshold or a critical value during a specific number of cycles. The threshold level and the number of cycles are generally pre-determined based on the industry standard or customer requirement (e.g., 80%~400 cycles). It usually takes quite a long time to complete the whole series of test cycles specified in the requirements; for example, 400 cycle-runs require at least fifty calendar days.



<Figure 2> Charging process in a lithium-ion battery system

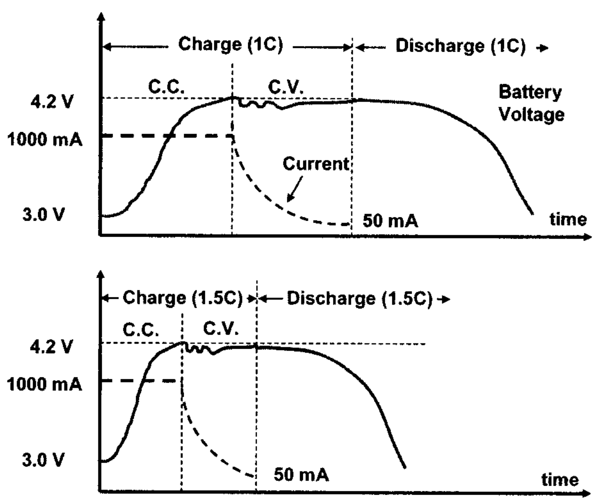
3. Types of Acceleration

In this section, we will briefly review three types of acceleration to shorten the test time and discuss how to apply these techniques to the cycle-life tests in the case application.

A simple scale acceleration failure time (SAFT) model [7] is assumed to extrapolate the lifetimes at the use condition from the test data obtained under each type of acceleration. The term scale-acceleration means that the failure time at a certain stress level can be obtained by simply multiplying its failure time at any other stress level by a time-scale factor (or an acceleration factor) [8]. Often, a single level of accelerated stress and a use-stress in an accelerated test set-up are loaded to check the validity of applied stresses in accelerating the same failure mechanisms that also occur in a normal environment. It is called a constant partially accelerated life tests (PALTs) [1]. For the PALTs, under the SAFT model, the acceleration factor can be easily obtained by calculating the ratio of the lifetimes at the use condition and the accelerated condition.

3.1 Usage-Rate Acceleration

The usage-rate acceleration method utilizes a higher than normal usage frequency. For an example, this type of accelerated test for certain shock absorber, used in cars, required to increase test frequency up to 600 cycles per hour, while a normal usage frequency is 200 cycles per hour. As a result, the test time could be reduced to three times [12]. In the case of lithium-ion batteries, the usage-rate acceleration method would increase the charge/discharge rate from 1C of the industry standard to 1.5C. The charge and discharge current of a battery is measured in C-rate and 1C means that a 1000mAh battery would provide 1000mA for one hour if discharged at 1C rate. Then, the require test time may be substantially reduced (See <Figure 3>).



<Figure 3> Charge/discharge profile (C.C. : Constant Current, C.V. : Constant Voltage) ; Top = 1C, Bottom = 1.5C

However, such advantage would be enjoyed only when the relating failure is caused by the same failure mechanism in both rates so that higher frequency do not produce any abnormal damage to the battery samples.

3.2 Stress Acceleration

Stress acceleration involves exposing the product to a harsher environment than the normal operating condition in order to expedite the failures, thereby reducing the required test time. Typical accelerating variables are temperature, voltages, mechanical load, thermal cycling, humidity, vibration, and etc. Failure times and/or censoring times at several accelerated stress conditions are then analyzed assuming a statistical lifetime distribution and a pre-specified stress-life relationship to estimate the product lifetimes at the use-condition. In this type of acceleration, type and the level of stress for accelerating the failure mechanism of a product should be identified in advance. Based on the analytical results of capacity degradation mechanisms in lithium-ion batteries, the following failure mechanisms have been identified with regard to the cycle life test as [3, 5, 10]:

- Interference in movement of lithium ion due to deposition of formation at negative electrode, which is produced by a reaction between minute amount of moisture in the battery and either electrolyte or lithium.
- Lowered capacity caused by unbalance in the party using an electrode catalyzing substance due to unbalance in current distribution inside the battery, which is caused by uneven distribution of electrolysis owing to charge/discharge cycle.
- Distortion in the structure due to repeated contraction and expansion of the lithium ion cell.

Bloom et al. [2] and Takei et al. [11] showed thermal stress could accelerate these mechanisms in their case applications.

3.3 Tightening Critical-Values

For some products, including light-emitting diodes (LED) and secondary rechargeable batteries, a failure can be defined as the time when the measured performance decreases to a predetermined critical degradation level that designates a non-functioning state or an incipient failure. In some contexts, this is referred to as a "soft failure." The lifetime of such a soft-failure type product obviously depends on the critical values and, if the degradation pattern is known ahead of test, then the tests can be further accelerated by tightening the critical values [14]. Then, the ratio between the times to reach each level may be interpreted as the acceleration factor in the SAFT model. Kitagawa et al. [6] classified the liquid crystal display (LCD) misalignment, which is the most significant failure mode of LCD consisting of watches and calculators, into three grades and suggested that time reduction may be possible for the testing of LCD through adjusting the grades, e,g., from the worst one to the mild one. For the lithium-ion batteries, if the acceleration factor is 2 between two critical values, 80% and 90% of the initial capacity, then the 400 cycle-runs can be reduced to the 200 cycles-runs with the 90% critical-value.

3.4 Data Analysis

In general, the accelerated test data are analyzed in the following framework.

① Fit lifetime distributions to the data at individual accelerated conditions. Here, the accelerated stress may be a specific usage-rate or environmental stress, sometimes critical value severer than the normal operating condition. The following Weibull and lognormal distributions are popular choices as the lifetime distributions:

$$F(t) = 1 - \exp\left\{-\left(\frac{t}{\eta}\right)^{\beta}\right\} \ ; \text{ Weibull}$$

$$F(t) = \int_{-\infty}^{t} \frac{1}{\sigma\sqrt{2\pi}} \exp\left[-\frac{(\log x - \mu)^2}{2\sigma^2}\right] dx$$

; lognormal

where $F(\cdot)$ is the cumulative distribution function.

② Check if the SAFT model is valid over the entire range of test conditions. This can be done by checking that the fitted lifetime distributions at each test condition have the same shape parameter (β) for Weibull and scale parameter (σ) for other distributions. Probability plots or the likelihood ratio test [9] can be used to check the model assumption. Specifically, the following statistics is calculated in the likelihood ratio test:

$$T = 2 \times \left(\sum_{i=1}^{l} \hat{L}_i - \hat{L}_0 \right)$$

where L_0 is the maximum log likelihoods with a common shape or scale parameter and L_i is the maximum log likelihood obtained by fitting a separate distribution to the data from each of the l accelerated conditions. If T is equal to or less than $\chi^2(1-\alpha; l-1)$, the $100(1-\alpha)$ percentile of the chi-square distribution with $l-1$ degrees of freedom, then the shape or scale parameters do not differ significantly at the level.

③ If the hypothesis test shows that the SAFT model is valid, then repeat fitting the lifetime distributions at each test condition under the assumption that the shape or scale parameters are all identical. Finally, calculate the acceleration factor using the estimated lifetimes. For an example, when the Weibull distribution is assumed, an acceleration factor can be computed as follows.

$$AF = \hat{\eta}_U / \hat{\eta}_A$$

where $\hat{\eta}_U$ and $\hat{\eta}_A$ are estimated Weibull scale parameter (characteristic life) at the use and accelerated condition, respectively.

4. Test Results

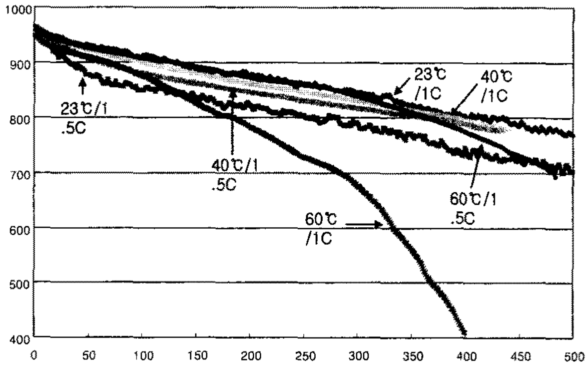
The accelerated test for lithium-ion batteries was planned as shown below.

The 23℃, 1C charge/discharge condition is presently being used by manufacturers. The highest temperature setting determined was 60℃ based on studies previously done by Bloom et al. [2] and Takei et al. [11]. The highest setting selected in charge/discharge speed was 1.5C. This was chosen upon consultation with a battery design expert, who identified it to be the highest adequate level. Since lithium-ion battery produces a lot of heat during charge/discharge cycle, there would be a gap in ambient temperature and actual temperature inside the battery, posing the danger of explosion. Since this could cause the failure mechanism different from that at the normal condition, a pre-test was performed to confirm the level of heat at the highest setting of 60℃. The temperature was measured in 1C and 1.5C rate. To measure temperature a thermal couple, attached to the surface of the battery, was used and it was located in a constant temperature chamber set at 60℃ ambient temperature. The results showed that the temperature increases by a maximum 6℃ with 1.5C, and maximum 4℃ with 1C. Since no trouble was reported up to 70℃ [2], we determined that it would be safe to conduct the test at 66℃, the highest temperature condition.

By monitoring the capacity during repeated charges/discharges under the test conditions in <Table 1>, the degradation trend of the battery capacity was obtained as the cycle proceeds (see <Figure 4>).

<Table 1> Accelerated test plan

	23℃	40℃	60℃
1C	8Ea	8Ea	8Ea
1.5C	8Ea	8Ea	8Ea



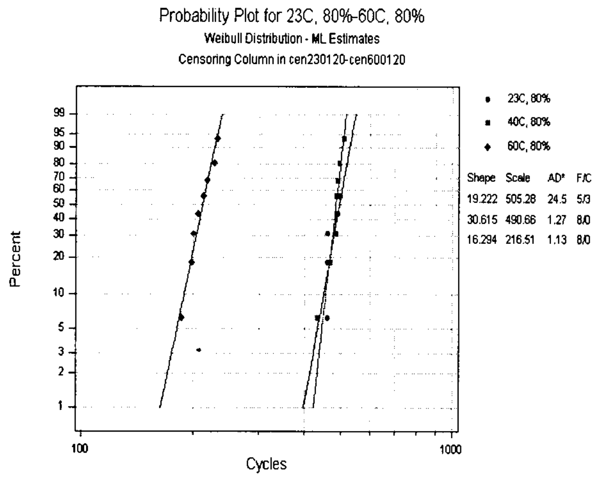
<Figure 4> Degradation trend of battery capacity

4.1 Assessing the Usage-Rate Acceleration

As shown in <Figure 4>, for 1C rate, the capacity decreased more rapidly at the higher temperature as expected. However, this is not general observation in this case application because the amount of degradation versus the temperature did not change coherently. In particular, the capacity degraded consistently at (23℃, 1C). On the other hand, the capacity at (23℃, 1.5C) dropped abruptly during the initial test, and then degraded consistently after a certain point in time. According to the engineer's opinion, abrupt decrease of the capacity during the initial test might be observed due to reduction of Li at the electrode side caused by high-rate charging. This type of failure mechanism is expected at the normal operating condition, (23℃, 1C). This leads to the conclusion that the SAFT model is not applicable for the usage-rate acceleration and all data from the 1.5C rate were not included in further analysis.

4.2 Assessing the Stress Acceleration

In order to check the validity of the SAFT model with respect to thermal stress, the failure data obtained under the conditions 23℃, 40℃, and 60℃ at 1C rate were analyzed. The critical value for failure was 80% of the initial capacity used as the conventional one. The data were plotted on a Weibull probability plot in <Figure 5> and Weibull distribution is found to be appropriate to describe the data because all the points in the plot were on straight lines. The figure indicates that the failure time at condition 40℃ was similar to that of condition 23℃; Consequently, the 40℃ is proven not to be enough to activate the effect of thermal stress on the lifetimes, which naturally follows by the reason of developmental design improvement of battery's cycle life. In details, there are two simultaneous mechanisms causing the capacity degradation over time. One is decreasing charge capacity with increase of cell impedance, and the other is losing Li source by irreversible reaction. Nowadays, methodology of appending the chemical additives, such as VC (Vinyling Carbonate), to the electrolyte of cells is broadly used by the battery manufacturers in order to prevent the side reaction which induces lost of Li source under high temperature. Therefore, it is natural that the batteries would not experience the degradation of capacity to the extent of moderately high temperature stresses such as 40℃.

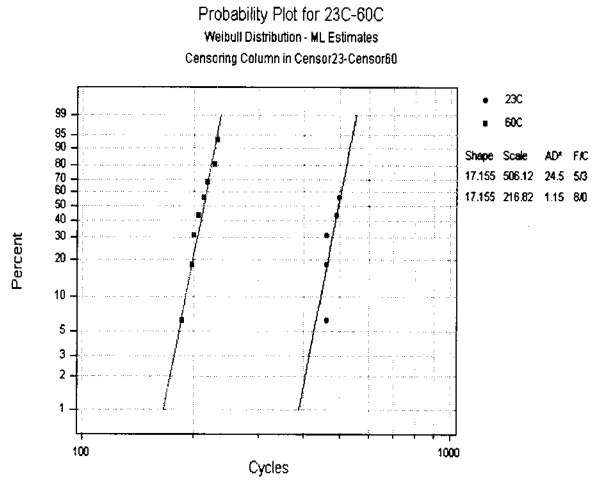


<Figure 5> Weibull probability plot of the failure time at each test temperature

In order to verify the acceleration model between 23℃ and 60℃, with the exception of condition 40℃ that did not show any acceleration effectiveness, the likelihood ratio test was performed at 5% significant level. The results from the likelihood ratio test are shown below:

$$T = 2[(-27.259-32.869)-(-27.3-32.887)]$$
$$= 0.118 \le 3.84 = \chi^2(0.95;1)$$

Since the assumption of the SAFT model holds between 23℃ and 60℃, we can fit the Weibull distribution with the same shape parameter for two test conditions as shown in <Figure 6>.



<Figure 6> Fitted Weibull distributions to the 23℃ and 60℃ assuming that the shape parameters are identical

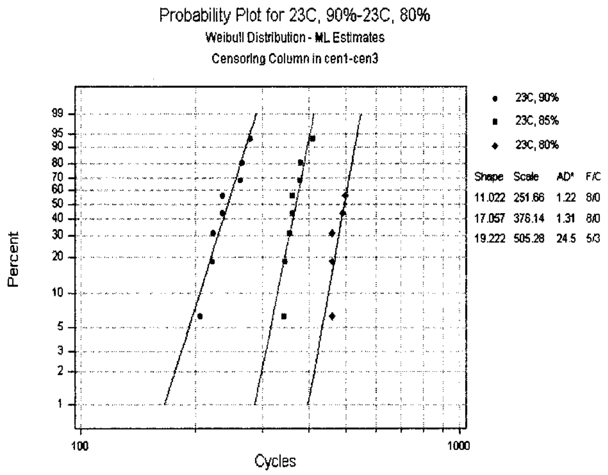
From the results, the acceleration factor between 23℃ and 60℃ was then calculated as 2.3 (≒ 506.12/216.82). With the acceleration factor, an activation energy can be estimated by modeling the Arrhenius relationship, which is a widely used model describing the effect that temperature has on the lifetime. The Arrhenius acceleration factor is [8]

$$AF = \exp\left[Ea\left(\frac{11605}{T_U} - \frac{11605}{T_A}\right)\right]$$

where Ea is the activation energy, T_A and T_U are accelerated and use temperature in the absolute Kelvin scale, respectively. Then, the activation energy is estimated by 0.195.

4.3 Assessing the Effect of Tightening Critical-Values

In order to assess the effect of tightening critical-values, we used the data obtained at 23℃/1C, the normal cycle life test condition. In this study, we picked up three critical values for assessing the acceleration: 80% (current), 85%, and 90%. Takei et al. [11] observed the capacity degradation pattern of the lithium-ion battery for an 8-mm video camera (type US 18650) under several accelerate conditions. Their test result indicated that 1500～2000 test data would be needed to minimize the extrapolation error when estimating 3500 cycles of nominal life for the case of CC-CV (constant current +constant voltage) used as a charging method of standard driving condition. Based on the fact that the nominal life of the product used in this study was about 500 cycles, it was determined that approximately 200 to 280 cycles of test data would be needed. The severest critical values was then set to 90% of initial capacity up to which the capacity is expected to degrade after the 200-280 cycle-runs. <Figure 7> is the Weibull probability plot of the failure data obtained under each critical value.

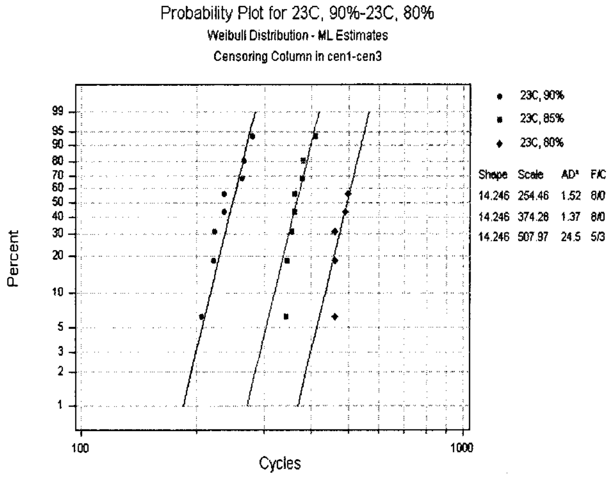


<Figure 7> Weibull probability plot for the critical values of 80%, 85%, and 90% of initial capacity

As shown in the figure, fitting the Weibull distribution to the data seemed reasonable. To evaluate validity of the SAFT model over the three conditions, the likelihood ratio test was performed at 5% significant level as follows:

$$T = 2[(-27.259 - 36.450 - 37.014) - (-27.521 - 36.678 - 37.505)] = 1.962 \leq \chi^2(0.95\,;\,2) = 5.99$$

Then, the scale parameters were estimated again to calculate the acceleration factors as shown in <Figure 8> by assuming that all of shape parameters in the Weibull distribution for the three critical-values are identical.



<Figure 8> Weibull probability plot for the three critical-values under assumption that the shape parameters are identical

For the case where the critical-value is adjusted to 90% of initial capacity, the acceleration factor can be computed as 2 (≒ 507.97/254.46). Therefore, the current cycle-life test with the 80%～400cycles could be replaced with a new one with the 90%～200cycles.

<Table 2> Measured capacity at 200 and 400 cycles

Sample ID	400cycle capacity	200cycle capacity	Sample ID	400cycle capacity	200cycle capacity
A10-3-05	87.676%	93.812%	E09-3-03	87.036%	92.667%
A13-1-02	87.523%	93.794%	E10-3-02	85.602%	91.292%
B08-1-3	85.599%	92.813%	E11-2-08	87.106%	92.369%
B14-1-4	86.243%	92.582%	F10-1-02	85.068%	90.387%
B15-1-2	86.087%	93.068%	F11-3-01	85.777%	91.346%
D04-2-6	87.500%	92.724%	F12-1-12	85.299%	91.488%
D06-1-06	87.763%	93.132%	F13-1-04	86.308%	91.951%
D07-1-09	87.769%	92.450%	F15-2-09	85.742%	91.125%
D08-1-04	86.676%	92.606%	G01-1-11	85.621%	90.775%
D09-2-08	86.959%	92.736%	G02-1-06	85.095%	90.473%
D11-1-2	86.024%	91.953%	G03-2-08	84.534%	91.455%
D22-2-5	83.724%	90.875%	G04-1-04	85.220%	91.026%
D32-2-5	84.363%	91.609%	J07-1	86.135%	92.615%
E01-1-02	85.073%	93.279%	J08-1	84.158%	91.478%
E03-1-03	85.846%	91.852%	K04-1	84.660%	91.943%
E04-2-11	84.274%	92.388%	K05-1	84.784%	92.005%
E05-2-09	83.853%	92.696%	K09-1	83.367%	90.860%
E06-1-1	85.068%	92.658%	K10-1	83.784%	91.461%
E07-3-4	85.485%	92.695%	K11-1	86.333%	92.785%
E08-1-06	84.062%	92.726%	L02-1	86.563%	93.227%

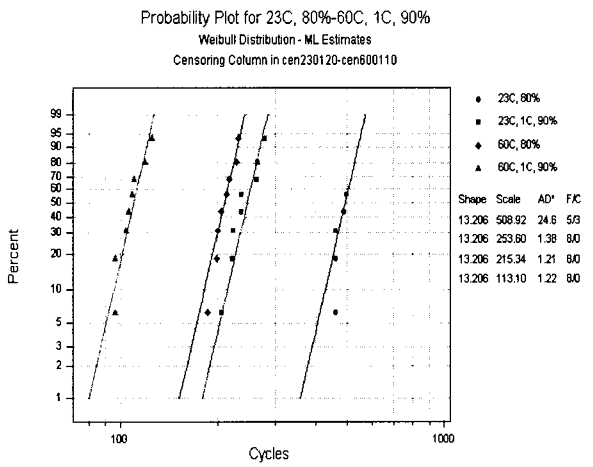
To check the validity of such replacement, both test standards were applied to the exiting data gathered for about one year at some manufacturing site (a total number of samples is 134), a part of which is shown in <Table 2>. As expected, both standards produce the same results on the conformity of each sample to the quality requirement.

4.4 Thermal-Stress Acceleration coupled with tightened critical-values

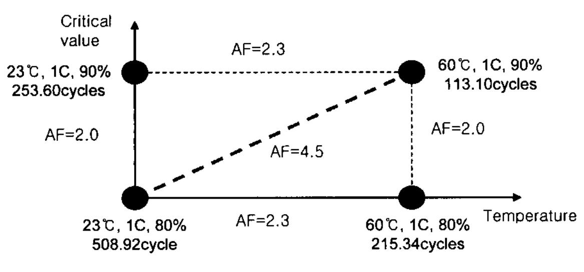
To evaluate the effect of the thermal stress acceleration when coupled with tightened critical-values, the likelihood ratio test at 5% significant level was first performed to check whether the SAFT model holds for the 4 combinations : 23℃/80%, 23℃/90%, 60℃/80%, 60℃/90%. The result was

$$T = 2[(-27.259-37.014-32.869-29.857) \\ -(-27.658-37.25-33.132-29.942)] \\ = 1.966 \le 7.815 = \chi^2(0.95,3)$$

and the relating distribution fit was obtained in <Figure 9>.



<Figure 9> Weibull plot with the same shape parameter assumed for each 4 combination



<Figure 10> Acceleration factors among the 4 combinations

<Figure 10> shows relationships among the four combinations in terms of the acceleration factor. The results show that the test time required for the current cycle-life test can be reduced at a minimum of 89 cycles (= 400/4.6) when employing the 60℃ test with the critical value of 90% of initial capacity.

5. Conclusions

This study proposes the accelerated methods to reduce the cycle-life test time of lithium-ion battery. Usage-rate acceleration, stress acceleration, and tightening critical-values were applied as acceleration methods for the batteries. The usage-rate acceleration with higher charge/discharge rate was proven not to work properly because the high-rate charge causes a little different failure mechanism at the accelerated condition. In the temperature acceleration method, the acceleration model is valid between 23℃ and 60℃, and the acceleration factor was estimated as 2.3. On the other hand, the acceleration factor of 2 can be obtained by tightening the critical-values from 80% to 90% of initial capacity. Further, the maximum acceleration factor obtained by combing the thermal stress with tightened critical value was 4.5, which leads to a substantial reduction from 400 cycles which is currently used as manufacturing standard to 89 cycles.

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