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Deterioration estimation of lithium-ion cells in direct current power supply systems and characteristics of 400-Ah lithium-ion cells

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ABSTRACT

Our evaluation of various characteristics of large-capacity 40 Ah and 80 Ah Lithium-ion (Li-ion) cells developed for industrial use shows that these cells exhibit reduced voltage characteristics and increased internal resistance in degraded conditions. We constructed a DC power supply system for telecommunications equipment using 80 Ah Li-ion batteries incorporating a function to estimate the cell status from the voltage characteristics during an automatic discharge of the battery. The Li-ion batteries in this system are maintained by a floating charge method, and we miniaturized the battery unit. In a field trial test lasting approximately 2 years, this DC power supply system performed well inclusive of the automatic discharge. The specifications and characteristics of prototype 200 Ah and 400 Ah cells, which were assembled to expand the application areas of Li-ion cells, are shown. Because these cells have similar characteristics to those of current 40 Ah and 80 Ah cells, they can be widely used as a substitute for conventional valve-regulated lead-acid (VRLA) cells.

1. Introduction

Lithium-ion (Li-ion) cells have high energy density and have been used to power a wide range of portable appliances. Large-capacity Li-ion cells with high energy density characteristics have been developed, and pre-production has progressed on industrial Li-ion cells that have sufficient capacity to serve as backup power supplies for telecommunications equipment [1]. Considering the background of these technical developments, we evaluated various characteristics of industrial Li-ion cells to clarify if they have adequate performance for use as backup batteries in telecommunications power supply systems and to identify the battery management functions that are required to keep the cell characteristics stable for long periods in such applications [2]. In backup batteries for telecommunications equipment, the cells need to be kept fully charged, and if possible, an automated system for doing so should be incorporated into the power supply system. Therefore, we clarified the relationships between the capacity and various parameters for these industrial Li-ion cells using deteriorated cells under high-temperature conditions. We then constructed a DC power supply system including Li-ion cells equipped with an automatic battery discharge function, leading to the establishment of this sort of automatic capacity estimation, and we conducted field tests at a base transceiver station providing actual telecommunications services.

Conventional DC power supply systems for telecommunications equipment use valve-regulated lead-acid (VRLA) batteries with a rated capacity around 5000 Ah, which is considerably more than the industrial Li-ion cells that are currently available. Therefore, we also fabricated large-scale Li-ion cells with a capacity of 200 Ah and 400 Ah per cell to expand their range of applications, and we evaluated the characteristics of these cells.

2. Industrial Li-ion cells

2.1. Industrial Li-ion cells and their basic characteristics

The industrial Li-ion cells we used to evaluate various characteristics and to construct the power supply system in this study are shown in Fig. 1, and the basic specifications of these cells are listed in Table 1. The cells are configured with positive and negative electrodes that are rolled up together with polyethylene (PE) separator sheets and accommodated in a metal container with a carbonate-based organic electrolyte. In these cells, lithium manganese oxide ($LiMn_2O_4$) spinel and graphite are used as positive and negative active material, respectively. $LiMn_2O_4$ was selected because of its higher thermal stability compared to that of $LiCoO_2$. The shape of the cell is rectangular to improve the thermal radiation and to reduce the space needed for installation. The energy density of these cells is approximately 2.5 times that of conventional VRLA

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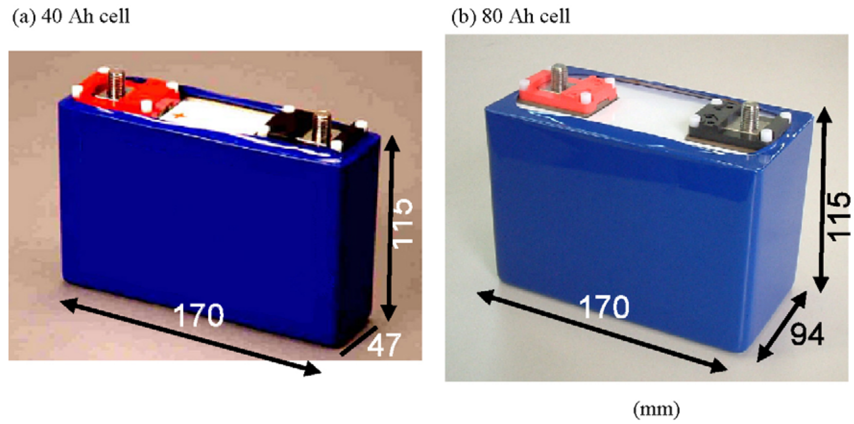

Fig. 1. Industrial Li-ion cells.

Table 1
Li-ion cell specifications

	Cell model	
	40	80
Nominal capacity (Ah)	40	80
Nominal voltage (V)	3.8	3.8
Dimensions (mm) (LWH)	170-47-133	170-94-133
Weight (kg)	2.1	4.1
Energy density (Wh kg^{-1})	72.4	74.1
(Wh l^{-1})	143.0	143.0

cells. A comparison of energy densities of VRLA and Li-ion cells is given in Fig. 2.

2.1.1. Charge characteristics

Fig. 3 shows the charge characteristics of these cells under constant current/constant voltage (CCCV) charge conditions at various charge voltages. Like lead-acid cells, these Li-ion cells exhibit favorable charging characteristics. The time taken to reach the preset charging voltage varies depending on the charge current at the initial stage of constant charging, but discharged electricity is almost fully restored after about 10 h of charging, even after a full discharge.

These Li-ion cells can thus be charged using CCCV charging methods, and they can be operated using the same float charging methods as conventional DC power supply systems for telecommunications equipment. As the −48 V DC power supply system supplies power with an output voltage at -48 ± 5 V, the floating charge voltage of the Li-ion batteries can be determined in this voltage range. The selection of the floating charging voltage is important for using Li-ion batteries in such DC power supply systems. In selecting the charge voltage, we must consider the battery composition (number of series-connected cells), off-load voltage, and available electricity from the cells that are governed by the off-load voltage, and in addition, the amount of electricity charged by the voltage. On the basis of our assessment regarding the effects of battery composition and charge voltages, we concluded that a battery comprising 12 cells and 4.1 V cell^{-1} are reasonable and acceptable for the DC power supply systems. Thus, we selected 4.1 V cell^{-1} as a standard float charging voltage for these Li-ion cells.

2.1.2. Discharge characteristics

Next, we will discuss the constant-current discharge characteristics. Fig. 4 shows the discharge characteristics of these Li-ion cells, where the terminal voltages over 3.5 V were obtained except in the final discharge period, at a discharge current rate between 1 h and 10 h. Also, the amount of electricity discharged with a 10-h discharge current is 10–20% greater than the nominal capacity, and this discharge capacity was almost the same below a 1-h rate (1 CA). In VRLA cells, the amount of discharged electricity decreases as the discharge current increases, especially over 0.6 CA. However, in the Li-ion cells, the discharged electricity is not influenced by the discharge current, so features such as size and weight reduction are especially promoted in applications where discharge current rates are high.

2.2. Deterioration of Li-ion cell and changes in their characteristics

For batteries used in backup applications, having not only a long lifetime but also some parameters or relationships that

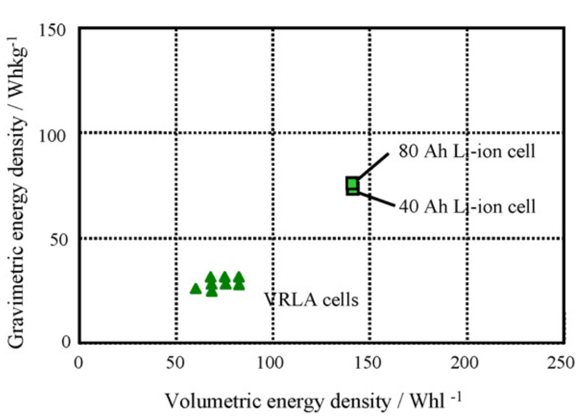

Fig. 2. Comparison of energy density of VRLA and Li-ion cells.

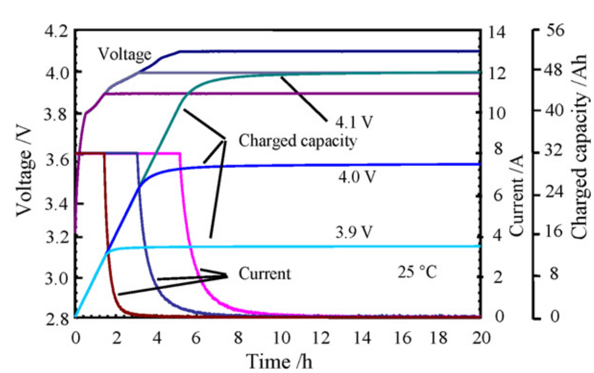

Fig. 3. Charge characteristics of Li-ion secondary cells.

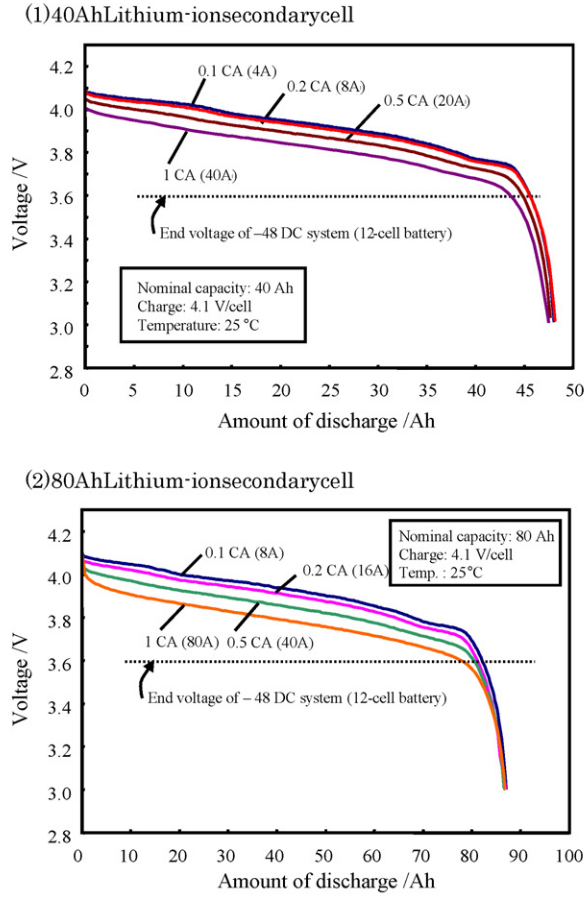

Fig. 4. Discharge characteristics of Li-ion secondary cells.

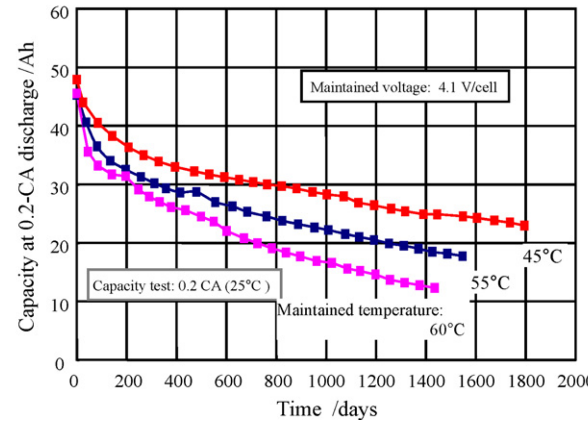

Fig. 5. Capacity changes of samples during high-temperature acceleration tests.

reflect the cell status during use are desirable. Therefore, we studied the relationship between the capacity and characteristics of our Li-ion cells using deteriorated cells that degraded under high-temperature accelerating conditions. The samples were 40 Ah Li-ion cells that had been subjected to continuous trickle charging ($4.1\,V\,cell^{-1}$) in high-temperature atmospheres of 45 °C, 55 °C, and 60 °C with periodic capacity tests and internal resistance measurements at 25 °C. After the capacity tests, recovery charging was performed at $4.1\,V\,cell^{-1}$, and then trickle charging was continued once again in the high-temperature atmospheres.

2.2.1. *Changes in capacity*

Changes in the capacity of 40 Ah cells degraded under high-temperature acceleration conditions are shown in Fig. 5. Capacity decreased greatly in the early stages of the tests. After that, capacity declined comparatively slowly, and an abrupt capacity decline did not occur under any condition throughout the tests. The temperature influenced capacity-reduction profiles, and the time the capacity took to decrease to a certain level also decreased at higher temperatures, thereby confirming the accelerated degrading effects of temperature. In general, the deterioration of Li-ion cells and decline in capacity were mainly caused by the formation of a solid electrolyte interface (SEI) layer on the negative electrode surface [3–6], and decomposition of electrolyte was mentioned as another cause of deterioration. It is reported that the growth of the SEI layer progressed gradually during use and capacity reduced in accordance with the thickness of the SEI layer. In this condition, capacity reduction/loss is expressed by the following equation [4]:

$$\text{Capacity loss} = kf\,t^{1/2} \quad (1)$$

where kf is rate constant and t is time.

Then we drew a figure of capacity changes on the basis of the relationship described by Eq. (1). Fig. 6 shows the capacity loss dependency on the square root of the test time. We found two kinds of fitting lines in each test temperature. One was observed in the capacity range over about 70% of the initial capacity (area 1), the other was observed below about 70% (area 2). The line had a larger coefficient in the higher temperatures conditions; this means the areas of higher degradation in the cell. On the other hand, in area 2, the coefficients are small compared to those in area 1; this means the areas of lower degradation. These kinds of plural fitting lines are also found in the paper by Yoshida et al. [4] on trickle-charged cells under 35 °C. However, in their report, the coefficient of the two lines did not differ as much. The reason is thought to be that their test temperature of 35 °C was not high enough to produce a distinct difference. Contrary to this, our test temperatures of over 45 °C are though to be high enough to make the differences distinct.

Our experimental results suggested two kinds of degradation mechanism in the test cells depending on the capacity ranges of over and below 70% of the initial capacity. In past papers [1,3], these kinds of two fitting lines were not reported. One reason was that the degradation test period was not long enough for cell capacity

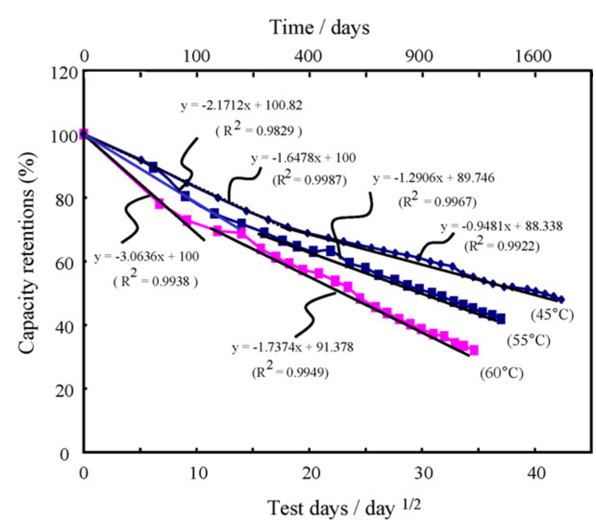

Fig. 6. Changes in discharge capacity retentions of 40 Ah Li-ion cells.

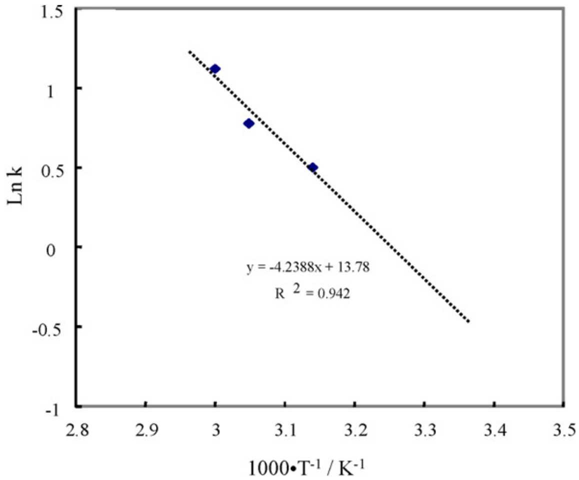

Fig. 7. Arrhenius plot of rate constant of step 1.

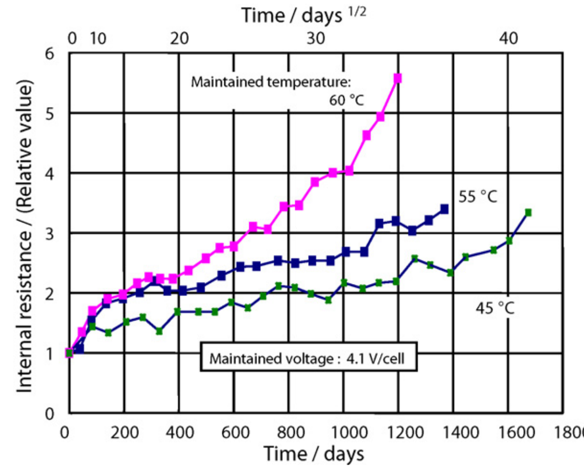

Fig. 9. Increase in internal resistance during acceleration tests.

to reach lower than 70% of the initial value. Additionally, the fitting lines in area 1 are thought to be useful to estimate the time for cells to reach a capacity of about 70% of the initial capacity after their use at any temperature, for example at 25 °C. Fig. 7 shows the Arrhenius plots of the rate constants obtained in area 1. The extrapolation of the fitting line gives a rate constant at any temperature required. For example, using the extrapolated rate constant at 25 °C, the time the cell capacity takes to reach 70% of the initial capacity can be calculated to be approximately 6 years. The time for cells to reach a lower capacity than 70% can be also estimated by additionally using the rate constants in area 2.

The variation in discharge curves of sample cells subjected to continuous trickle charging at 45 °C is shown in Fig. 8. As the test progressed, the discharge time decreased, and the capacity gradually decreased. The discharge voltage curves also changed during this test, and in cells with reduced capacity, cell voltages were clearly reduced at any cut after the beginning of the discharge.

The change in internal resistance during these tests is shown in Fig. 9. As time passed, the internal resistance increased, and at higher temperatures in particular, internal resistances were higher at any cut after the tests. This behavior corresponds to the profiles of reduction in capacity with respect to time. When we take a look at the precise behavior of the internal resistances, we can see

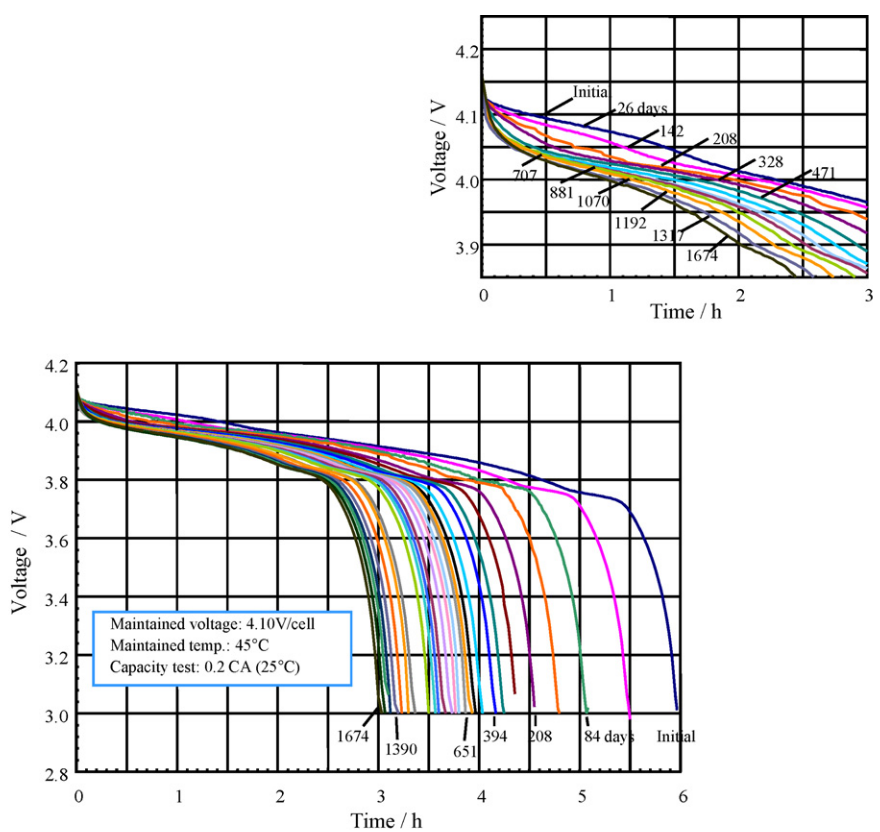

Fig. 8. Discharge characteristics of Li-ion cells during tests.

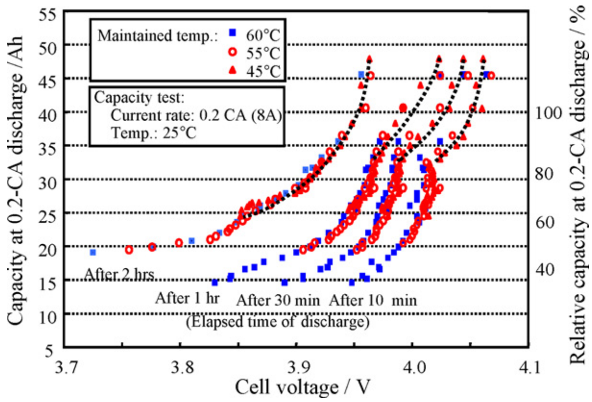

Fig. 10. Capacity vs. voltage relationships of samples that deteriorated under various conditions.

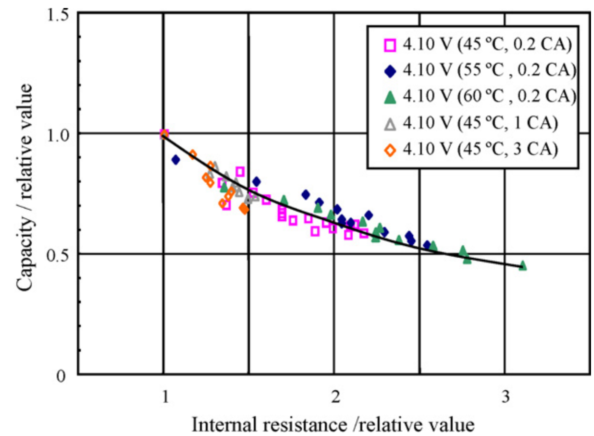

Fig. 11. Internal resistance vs. relative capacity.

they increased rapidly in the duration that corresponds with area 1, where rate constant was relatively high, and after that the increase ratio was reduced. This behavior can be observed especially in the sample degraded at 60 °C.

2.2.2. Relationship between residual capacity and cell parameters

We then studied the relationships between the discharge characteristics and the internal resistance and cell capacity obtained in the tests.

First, from the discharge characteristics of the Li-ion cells subjected to accelerated degradation at each temperature shown in Fig. 8, we clarified the relationship between cell residual capacity and cell voltages at arbitrary elapsed times. Fig. 10 shows the relationship between the residual capacity and the cell voltage at arbitrary elapsed times during discharge. A significant correlation between the cell residual capacity and the cell voltage was found, regardless of the accelerated degradation temperature. We thus inferred that high temperature acceleration caused degradation in these Li-ion cells. In this way, we found a correlation between the residual capacity and the cell voltage at any time elapsed during a constant current discharge in Li-ion cells used in this work. Thus, even in DC power supply systems, the residual capacity of the Li-ion cells can be estimated from the cell/battery voltage measured during a brief discharge at a constant current.

We also examined the relationship between the cell capacity and internal resistance. As Fig. 11 shows, the results indicate a good correlation between the two that demonstrates the residual capacity can be estimated from the internal resistance. Thus, if equipment for automatically measuring the internal resistance is installed in DC power supply systems, then this relationship should enable us to estimate the cell capacity. However, measuring the internal resistance of cells generally requires expensive high-precision equipment.

Thus, we gave priority to a method using a discharge function to ascertain the battery state from voltage measurements in a DC power supply system used in field tests, described next.

3. DC power supply system

3.1. Overview of power supply system

The DC power supply system produced for tests in the field trials has a backup function, which was configured from a storage battery unit and a power conversion unit including rectifiers and a control unit. The configuration of the system and an illustration of it are shown in Fig. 12. The upper stage is the housing for the power converter, and the lower stage is the housing for the storage battery. The storage battery can contain two 80-Ah Li-ion batteries in parallel;

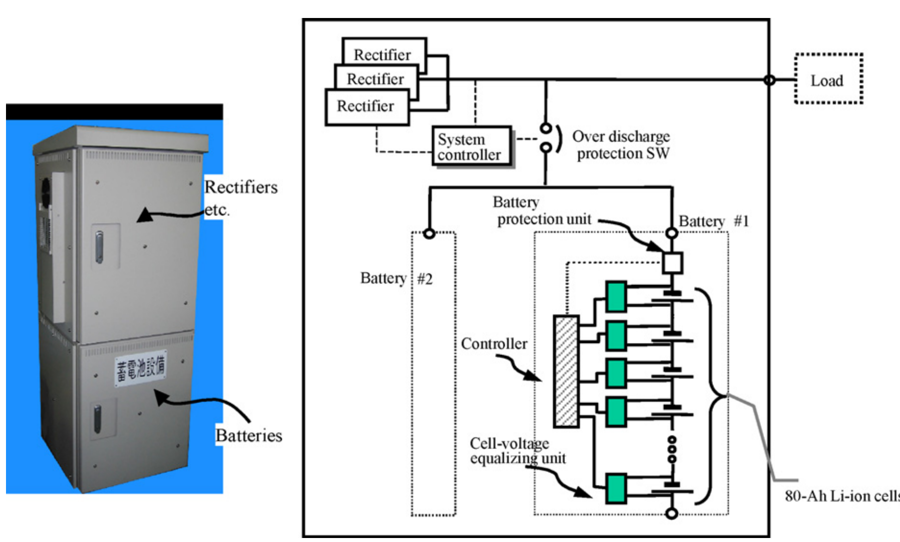

Fig. 12. DC power supply system.

Table 2
Specifications of DC power supply system for field test

	Model	
	Conventional	Developed
Battery type	VRLA	Li-ion
Battery model	12 V-65 Ah	80 Ah
Battery composition (number of cells)	4 series × 4p (24)	12 series × 2p (12)
Total capacity (Ah)	6240	1920
Output voltage (V)	−54.6	−49.2
Output current (A)	30	30
Charge method	Floating charge	Floating charge
Backup time (h)	6	6[a]
Size (L/W/H) (mm)[b]	700/750/1680	700/750/1680
Total mass (kg)	802	414
Rectifier, case	269	269
Battery	432	98
Battery controller	–	47
Battery box	101	–
Battery volume (L)	163	51

[a] Based on actual capacity.
[b] Exclusive of battery box.

the batteries are each composed of 12 cells connected in a series. In the figure, the configurations of the Li-ion battery and the DC power supply system are also shown schematically. The battery is connected in parallel with the load at the output of the rectifier, and it is constantly kept at a fully charged state by the floating charge method. Because the battery is configured with an integral control unit, each cell can be managed individually, and batteries can be installed/replaced or removed by connecting them to the rectifier outputs as necessary.

Table 2 compares the specifications of the DC power supply system used in the field tests with those of the conventional system. The rated output voltage of the system was set to 49.2 V considering the optimal charging voltage of the Li-ion cell of 4.1 V cell^{-1} and the number of cells in the battery. This value is about 5 V lower than the 54.6 V voltage of conventional power supplies but is adequate for the required voltage of the load. The maximum output current of the DC power supply system used in the field tests is 30 A, and the backup time of the storage battery is 6 h. In a conventional power supply system equipped with VRLA batteries, four sets of four series-connected 12 V/65 Ah VRLA batteries are used in parallel. An additional battery enclosure is used outside the DC power supply main unit to cope with these batteries. In addition, the same discharge time is compensated with two parallel sets of 80 Ah batteries. In this way, the number of cells that are used in a serial/parallel configuration of Li-ion batteries is reduced. Consequently, the total cell capacity of batteries is reduced to 1920 Ah, which is about one third that of VRLA batteries (6240 Ah). As a result, the net volume of the battery is also about one third that of the VRLA battery, while the weight is reduced to approximately one quarter. Because a battery control unit is required in Li-ion batteries, the weight of the control unit (47 kg for two units) is also added to the battery. However, even if this weight increase is taken into consideration, the overall weight of the battery is reduced to approximately one-third. The end result is that the total weight of the system is reduced by about half due to the substantial reduction in the weight taken up by the VRLA battery.

3.2. Cell management and battery configuration

In general, when Li-ion cells are used as batteries by connecting single cells in series, they must retain adequate performance and safety over long periods of time. Therefore, each Li-ion battery we used in our DC power supply system is equipped with a dedicated control unit and cell-voltage equalizing circuits, and functions were provided for keeping the cell/battery safe inclusive of cell-voltage management and adjustment to an appropriate voltage.

(1) Cell voltage management and voltage equalization
The voltage of each individual Li-ion cell in the battery was kept at a charge voltage suitable for maintaining the cell's capacity and lifetime. For this purpose, a cell-voltage equalizing circuit that is provided in parallel with each cell bypasses the excess charging current when the voltage exceeds a set value. In our system, the standard adjustment target value was determined to be 4.1 V cell^{-1}. However, for actual operation, we revised this value by considering the change in the rectifier output voltage.

(2) Single-cell protection
- Prevention of overcharging and over discharging: To prevent overcharging and over discharging from occurring, the cell voltages are monitored, and the batteries are disconnected from the system. The charging/discharging is stopped when the system detects either an excessive increase or decrease in the cell voltage during charging or discharging, respectively, in any cell.
- Prevention of elevated temperatures: When a cell is detected to reach a temperature above a prescribed value, the batteries are disconnected from the power line to prevent charging/discharging of the battery.

(3) Detection of control unit malfunctions
When a malfunction occurs in a control unit, the batteries are disconnected and the charging is stopped.

(4) Automatic operative functions
This DC power supply system is equipped with two functions: one is for disconnecting the battery from the load during discharge when the battery voltage falls at a preset value to prevent excessive discharging of the cells, and the other is a periodic battery discharge by automatically dropping the rectifier output voltage within the working voltage range of the load. The latter function is useful to check the health of the DC power supply system during actual operating conditions as well as the state of health of the battery. We measured the voltage characteristics of the Li-ion batteries in the DC power supply system when it was discharged to the real load by using the function for automatically dropping the rectifier output voltage.

3.3. Field test results

We installed the DC power supply system at a base transceiver station providing actual telecommunications services, and in June 2006, we began field tests during actual service conditions. The test site is located in western Japan (northern Kyushu), where average temperatures are high and where many typhoons occur in summer. The test system is shown in Fig. 13. A measuring system was installed beside the DC power supply main unit, and various kinds of measurement data were collected from the DC power supply system during the tests and transmitted via a communication line to our R&D center in Tokyo. Any alerts generated in the power supply system can also be transmitted to our Tokyo center.

3.3.1. System stability

The system stability was checked by measuring parameters such as the rectifier output, the cell voltages, and current supplied to/from the battery while rectifiers were feeding power to the load continuously. The system has been operated in summer and winter since the start of the field test in June 2006, and so far, it has functioned well and has generated no alerts.

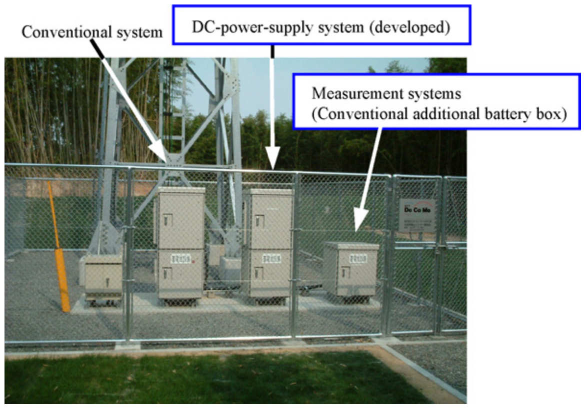

Fig. 13. DC power supply system at field test site.

Fig. 14 shows the changes in cell temperature and cell voltage during the test period. Because the environmental temperature influences the rectifier output, the output voltage varies depending on the ambient temperature. Consequently, temporal changes in the cell voltages occur as the temperature of the test site changes. However, the range of these voltage variations was within ±10 mV of the adjustment target voltage (4.10 V), and we found that the cell voltage equalizing circuit functioned well even during the actual test conditions.

3.3.2. Characteristics during automatic discharge tests

Fig. 15 shows the battery discharge characteristics resulting from the automatic discharge function incorporated into the DC power supply system. As shown in this figure, the battery was discharged due to a drop in the rectifier output voltage without any instantaneous breaks in the supply current, and the telecommunications system continued to function well. Also, the voltages and discharge currents of the two sets of batteries were well aligned during the discharge, and afterwards, the system switched over to recovery charging without any difficulty.

Fig. 16 shows the battery voltage characteristics during automatic discharge tests that were performed repeatedly during this field test. Although the cell temperature affects these discharge characteristics, the first and fourth characteristics, which were obtained at more or less the same temperature, show a similar discharge. Because the load currents were almost constant during the field test, we can infer from these characteristics that the Li-ion batteries installed in this DC power supply system did not undergo any large changes in their conditions. Thus, we are confident that the deterioration of Li-ion cells/batteries can be

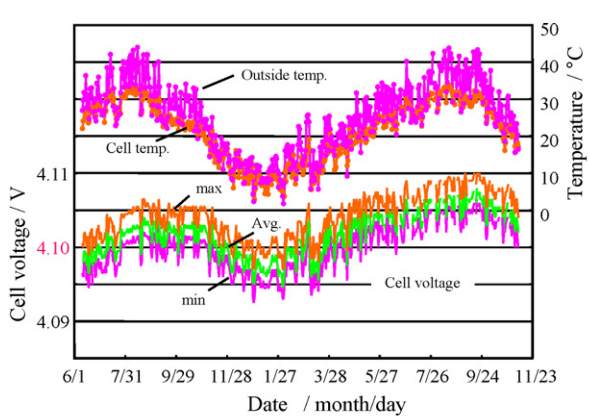

Fig. 14. Cell-voltage variations during field test.

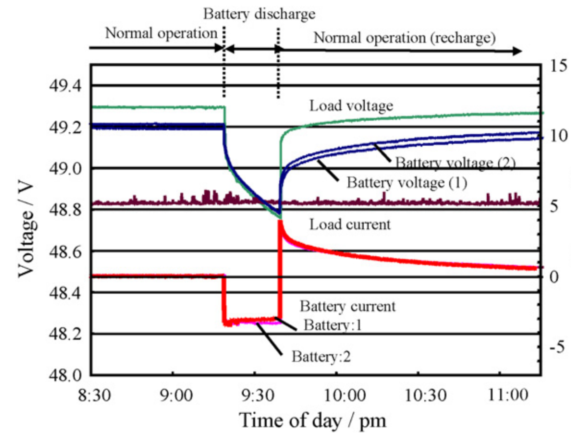

Fig. 15. System responses during automatic discharge tests.

estimated by monitoring the voltage in the DC power supply systems.

4. Development of Li-ion cells with larger capacity

4.1. Single-cell characteristics

We made prototype large-scale Li-ion cells with capacities of 200 Ah and 400 Ah to test the potential of these cells for industrial applications. Fig. 17 is a photograph of the prototype 400 Ah Li-ion cell. The cell is fabricated by enlarging components based on a similar design concept to that of the 40 Ah and 80 Ah cells. Active materials for positive and negative electrode are the same as that used in the 40 Ah cells. Specifically, the width and length of the electrode sheets were determined by taking into consideration the amount of active material required to store the electricity, and the cell-components were placed in a metal case. In these cells, electrode sheets with a width of approximately 300 mm were applied. Due to the cell design, the energy density was slightly higher than that of the current 40 Ah and 80 Ah cells.

Fig. 18 shows the discharge characteristics of the 400 Ah Li-ion cells. In the discharge current region from the 10-h rate (0.1 CA) to the 1-h rate (1 CA), the cell has a high voltage of over 3.5 V except during the final discharge stage, and the discharge capacity is more or less the same regardless of the discharge current. Similar characteristics were also obtained for the 200 Ah Li-ion cells, and these discharge characteristics were more or less the same as those of the

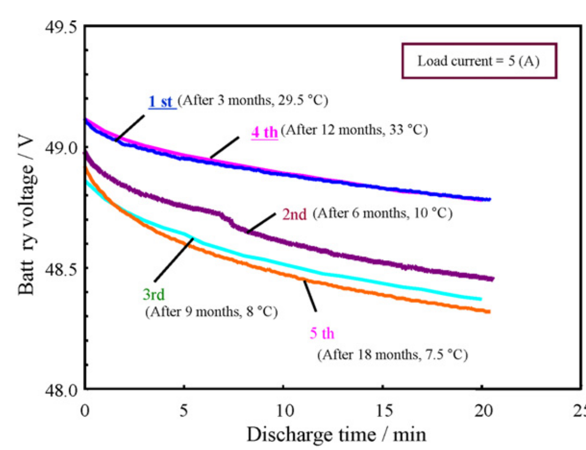

Fig. 16. Discharge characteristics of Li-ion battery in automatic discharge tests.

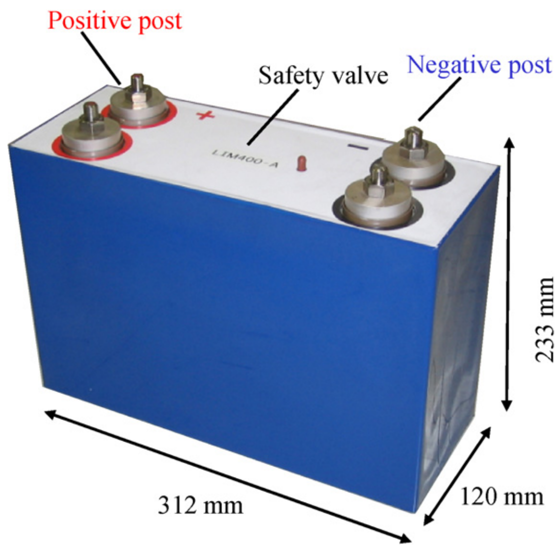

Fig. 17. 400-Ah Li-ion cell.

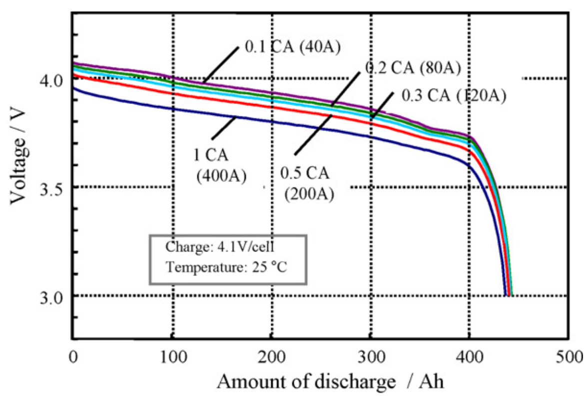

Fig. 18. Discharge characteristics of 400 Ah Li-ion cell.

Li-ion cells (40 Ah and 80 Ah cells) described earlier. Because these discharge characteristics contribute to reducing the battery size, the 200 Ah and 400 Ah Li-ion cells should also reduce the battery size to the same degree as the current Li-ion cells when the VRLA batteries are replaced with the Li-ion batteries.

4.2. Application range

Fig. 19 shows the range of loads to which the 200 Ah and 400 Ah Li-ion cells should be applied. In general, with a VRLA cell, the amount of discharged electricity decreases as the discharge current increases, dropping down to approximately 60% of the rated capacity at the 1 CA current. Thus, for example, a cell with roughly 1.7 times the rated capacity is required for back up batteries with a 1 CA load current. However, with a Li-ion cell, the discharged electricity remains more or less constant regardless of the discharge current. Accordingly, if we compare VRLA cells and Li-ion cells with the same rated capacity, then the Li-ion cell can handle larger load currents for the same backup duration, as shown in the figure. Thus, 200 Ah Li-ion cells can be substituted for 500 Ah VRLA cells at a backup time of 1 h, for example, and for a backup time of 30 min, they can replace

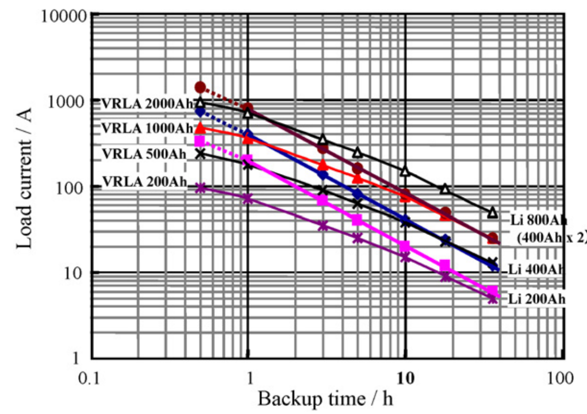

Fig. 19. Applicable range of Li-ion cells.

1000 Ah VRLA cells. Furthermore, 400 Ah cells can replace 500 Ah VRLA cells for over 10 h and replace 1000 Ah VRLA cells for around 1 h. This shows that Li-ion cells with capacities of 200 Ah and 400 Ah can be used as replacements for VRLA cells with capacities ranging from 1000 Ah to 2000 Ah. They are used in large quantities in the telecommunications power systems. And, as mentioned, because the number of Li-ion cells in a battery can be reduced to about half (12 cells) the number of cells in a conventional VRLA battery (24 cells), the total cell capacity installed in these large-capacity Li-ion batteries is much less than that of conventional VRLA batteries.

5. Summary

We studied the relationship between the capacity and characteristics of industrial Li-ion cells in deteriorated states, and we clarified that the capacity is related to the internal resistance and the cell voltage during constant current discharge. By focusing on the relationship of the capacity and voltage characteristics during discharge, we constructed a DC power supply system with automatic discharge functions, and we performed field tests. The use of Li-ion batteries enables substantial reductions in the size and weight of battery units in power supply systems. The cell status can be diagnosed by the automatic discharge test function in DC power supply systems based on the cell/battery voltages during a brief discharge.

We also fabricated 200 Ah and 400 Ah Li-ion cells, and we found that large-capacity Li-ion cells can be produced with similar characteristics to current 40 Ah and 80 Ah Li-ion cells, thereby demonstrating the potential of Li-ion cells as replacements for the VRLA batteries in telecommunications systems.

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