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Quick testing of batteries in lithium-ion battery packs with impedance-measuring technology

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Abstract

We discuss a rapid testing of capacity in Li-ion battery packs with impedance-measuring technology to evaluate their capability in mobile phones. Our measurements of the capacity and impedance at 1 kHz for various kinds of battery packs conclusively proved that there is a strong correlation between capacity and impedance. It can be applied even when the pack consists of not only a Li-ion battery but also a Positive Temperature Coefficient (PTC) device and a protection circuit in-series, and this correlation is largely unaffected by the degree of charge. The results we obtained from measuring impedance revealed the possibility of quickly assessing degradation in Li-ion battery packs.

Keywords: Capacity; Deterioration; Li-ion battery; Battery pack; Impedance

1. Introduction

Recently, mobile phones are changing from a second-generation (PDC, GSM) system gradually to a third-generation (W-CDMA) system throughout Japan. It is necessary to improve various hardware specification items in developing mobile phones and the most important one is operating time because they have sufficient talking and standby time. For this reason, developments in mobile phones have mainly been moving in two directions. The first has been toward low power consumption and the second has been toward the development of a high-power density secondary battery such as a Li-ion battery, which has higher output voltage and higher power density than either a Ni–Cd or a Ni–MH battery. The Li-ion battery has recently been widely used in mobile phones throughout Japan and have contributed to remarkable improvements of its operation time.

There have been various problems with the Li-ion battery. The main ones have involved safety and replacement. Care must be taken not to overcharge it and high temperature needs to be avoided because it contains active lithium material and electrolytes of organic carbonates. To ensure safety under normal use in a mobile phone, only one Li-ion battery (cell) has been built into a battery pack, along with other protective measures in the form of devices that have been used to protect it against overcharging, overdischarging, overcurrent, and high temperature. Consequently, the pack has been widely and safely used as a power source for mobile phones and mobile PCs.

In terms of frequency of replacement, the Li-ion battery is subject to cyclic discharge and recharge that causes it to gradually lose its battery capacity. It leads to a reduction in talk and standby time. However, the degree of degradation can currently only be judged from a feeling that talk time was not as long as it ought to be, particularly after the battery pack was fully charged, or from the residual capacity meter display on the phone.

The basic and most reliable technique for determining battery degradation involves conducting capacity testing in which a fully charged battery is discharged [1]. However, this test takes a long time to complete. Complex impedance (ac-impedance) has been studied over the long term as a means of analyzing the internal reaction in sealed lead-acid batteries (VRLA batteries), Ni–Cd batteries, and Ni–HM

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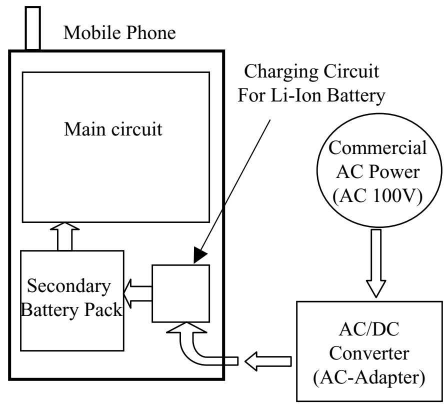

Fig. 1. Power system for mobile phones.

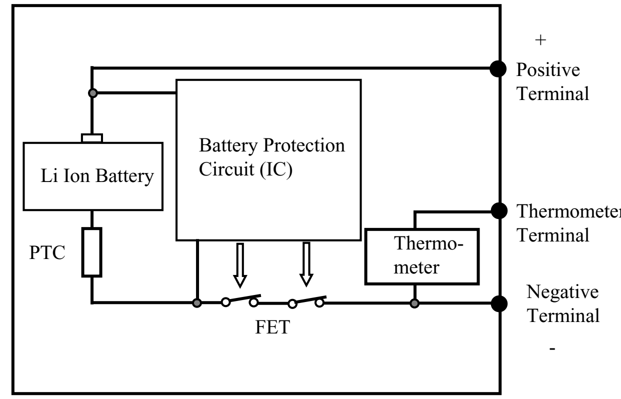

Fig. 2. Li-ion battery pack for mobile phones.

batteries, and has recently been adopted as a quick sway of evaluating battery degradation [2,3]. This method has most frequently been put to practical use in the large VRLA batteries that are widely used in telecommunications buildings. The logarithm for NiCd impedance has a linear relationship to its capacity. However, there have been no reports on the relationship between impedance and capacity degradation from the perspective of evaluating Li-ion battery packs with protective devices.

In this paper, we measured the ac-impedances of numerous kinds of battery packs for mobile phones and compared their relationships with deterioration in battery capacity. We used many deteriorating Li-ion battery packs, which were made like this by discharging and charging them over numerous cycles. The results we obtained from measuring impedance revealed the possibility of quickly assessing degradation in Li-ion battery packs.

1.1. Mobile phone and battery pack power systems

Fig. 1 shows the basic configuration for a power system in mobile phones. During charging, commercial 100 V power (ac) is converted to a dc voltage through an ac adapter (ac/dc converter), and the dc power is input to an Li-ion battery pack in the phone through a built-in charging circuit in the mobile phone.

Fig. 2 shows the configuration for a commercial battery pack, which is composed of a Li-ion battery, with $LiCoO_2$ as cathodic material and graphitized carbon as anodic material. There is also a protective IC, two-series Field Effect Transistor (FET) switches (circuit switches controlled by protective IC), a thermometer (which monitors the temperature in the battery pack), and a Positive Temperature Coefficient (PTC) device, which behaves like a reversible electric current fuse. The Li-ion battery is quite sensitive to overcharging, overdischarging, and high temperature. The packs have these devices built in to insure safety.

Fig. 3 shows the voltage and current characteristics during a cycle of charge and discharge for a Li-ion battery pack. During the charge period in the figure, the battery pack is first charged at a constant current and then the voltage rises. It is charged at a constant voltage after the battery voltage reaches a fixed value. During constant voltage charging, the battery current decreases gradually. Then, the battery pack charge stops when the current reaches the end of the charge current. The discharge voltage and current curve are also shown, during the discharge period, after charging has been completed. The voltage during discharge gradually decreases under constant current. Battery pack discharge stops when the voltage reaches the discharge end voltage.

We can see that the battery capacity of a Li-ion battery in a pack decreases gradually through repeated cycles of charge and discharge. Fig. 4 shows the change in voltage curves in a Li-ion battery pack due to capacity degradation over 800 cycles. Each curve was plotted by measuring after 1–800 charge and discharge cycles in steps of 50 cycles. Fig. 5 plots battery capacity for 20 samples from 1 to 800 cycles progress. From Figs. 4 and 5, it is obvious that the discharge time and battery capacity decrease as the number of cycles increases. In particular, more than half of the samples after 800 cycles has 50% or less of capacity compared with the new one. Therefore, it is clear that the degraded capacity depends on the number of cycles progress.

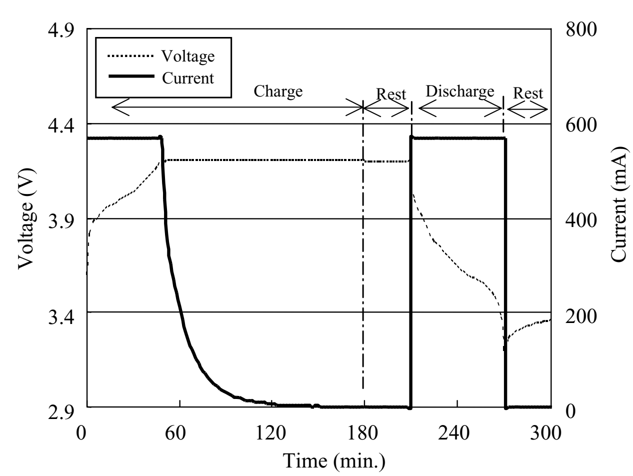

Fig. 3. Voltage and current curves for Li-ion batteries during charge and discharge.

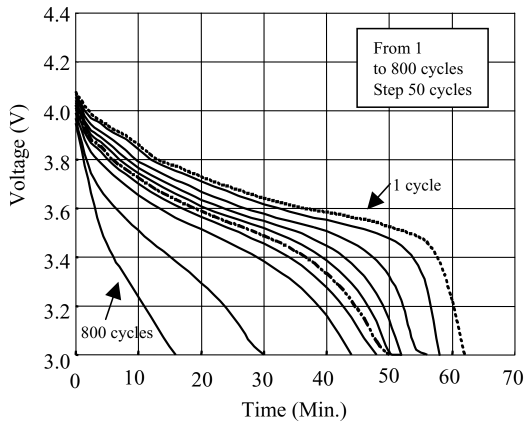

Fig. 4. Voltage curves for discharge characteristics.

1.2. Equivalent circuit for Li-ion battery pack and its impedance characteristics

The simplest equivalent circuit model for Li-ion battery packs in Fig. 2 is represented in Fig. 6 [4]. Here, Rs is internal resistance, which is based on a liquid electrolyte and the collector portion, and C1, Rp1 and C2, Rp2 are the

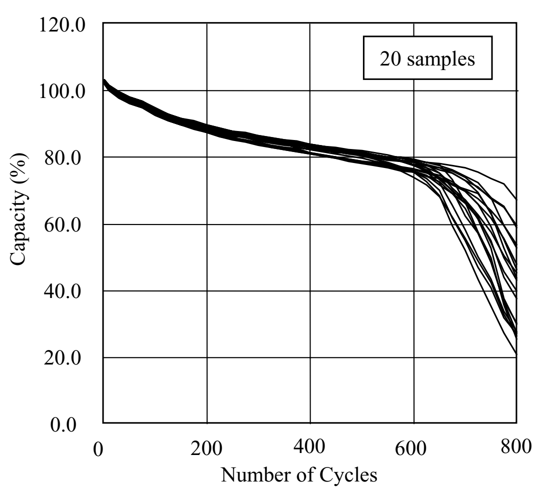

Fig. 5. Life cycle characteristics of battery capacities.

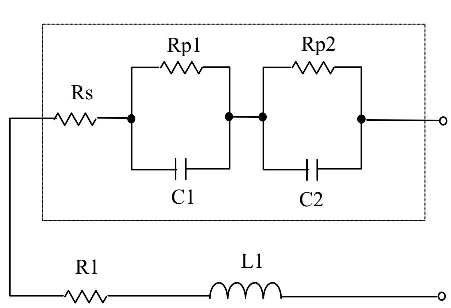

Fig. 6. Equivalent circuit for Li-ion battery pack.

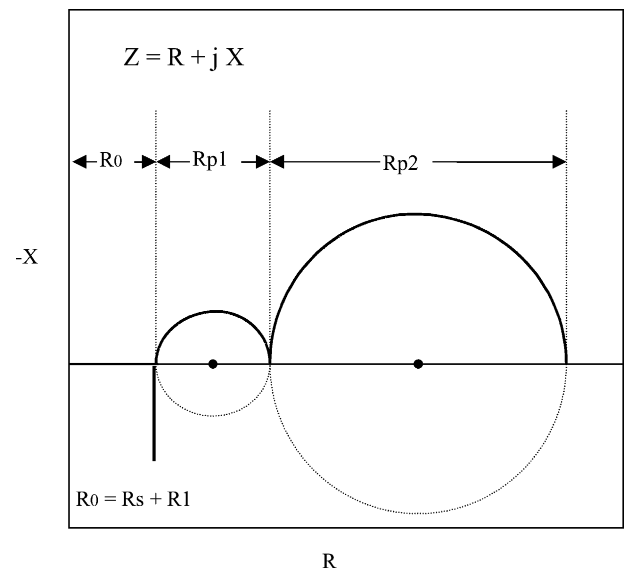

Fig. 7. Schematic Cole–Cole plot for Li-ion battery pack.

respective reaction resistance and capacitance components on both electrode–electrolyte interfaces. R1 is the sum of on-resistance in two-serial FETs and the resistance of a PTC, and L1 is the inductance, which is based on the wiring in the battery pack.

The complex impedance of an equivalent circuit in Fig. 6 can be expressed as Fig. 7, which is the schematic Cole–Cole plot of an Li-ion battery pack. In this plot, the horizontal axis is the real part of impedance (expressed as R), and the vertical axis is the imaginary part of impedance (expressed as $-X$). Generally, there are two half-cycles reflecting the reaction velocities at both electrode–electrolyte interfaces for a Li-ion battery. R0 in the figure expresses the sum of RS and R1, and Rp1 and Rp2 express the reaction resistance of the negative electrode and the positive electrode. In addition, the bold vertical line below the line is based on the inductance of the wiring.

2. Experimental

We measured impedance characteristics using Li-ion battery packs with various degradation capacities or under various charging conditions to clarify the relationships between battery capacity deterioration and battery impedance. We used the commercial battery packs made for mobile phones available throughout Japan in this experiment. The Li-ion battery (cell) is prismatic and its capacity is around 600 mAh. Normally, it is best to measure both electrode terminals of the battery (cell) directly in the pack to prevent the attached circuitry from affecting the original impedance of the battery (cell). However, the impedance of the battery pack can only be measured using the positive and negative terminals because the commercial battery pack has no other terminals.

2.1. Preparation of deterioration battery packs and measurement of impedance

We prepared numerous fresh and deteriorated mobile phone Li-ion battery packs for this experiment. There were 10 models (nine by Japanese companies) in these packs, and a total of 800 battery packs were used. Deteriorated battery packs were mainly prepared based on cycles of discharge and charge. Discharge and charge were repeated under conditions where the maximum discharge and charge current was 1 C mA (C means a number equivalent to the rated capacity of an Li-ion battery). The batteries were discharged to 3 V, and the maximum charging voltage was 4.2 or 4.1 V (depending on the charging specifications of each battery pack) for 3 h. Discharge and charge took place in an incubator where the temperature was fixed at 25 °C (near room temperature).

We used frequency response analysis equipment to determine the complex impedance (a system that combined the Solartron (UK) SI1287 and SI1253) which is widely used to measure the ac-impedance of a battery. We injected sine wave constant current, which is large enough to overcome noise and small enough not to charge the internal state of the battery, and analyzed the battery voltage response to calculate the impedance from a range of several 100 mHz to several 10 kHz. Using this method, we obtained a Cole–Cole plot of the Li-ion battery packs. However, we also used a simple impedance measuring instrument (the Hioki 3560 AC mohm Hitester) when we measured impedance that had a fixed frequency value over a short time.

2.2. Basic impedance characteristics of Li-ion battery packs

Fig. 8 shows a Cole–Cole plot of a fresh, fully charged Li-ion battery pack. Complex impedance data in the Cole–Cole plot was measured in a range from 100 mHz to 10 kHz. We can see two half-cycles in the Cole–Cole plot as mentioned for Fig. 7. The value at the intersection with the R axis ($-X = 0$), which is equivalent to R0, is the sum of the original RS of a battery (cell) and the R1 of protective devices in the battery pack.

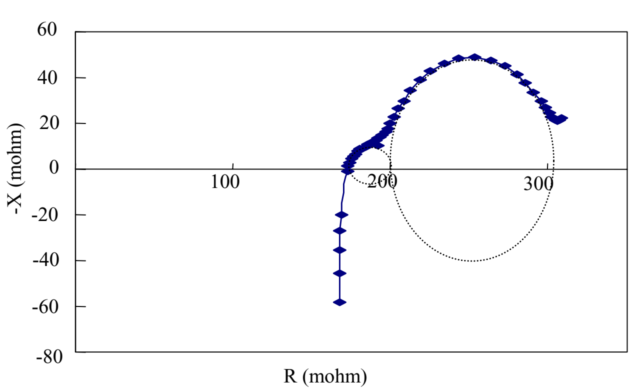

Fig. 8. Cole–Cole plot data for Li-ion battery pack.

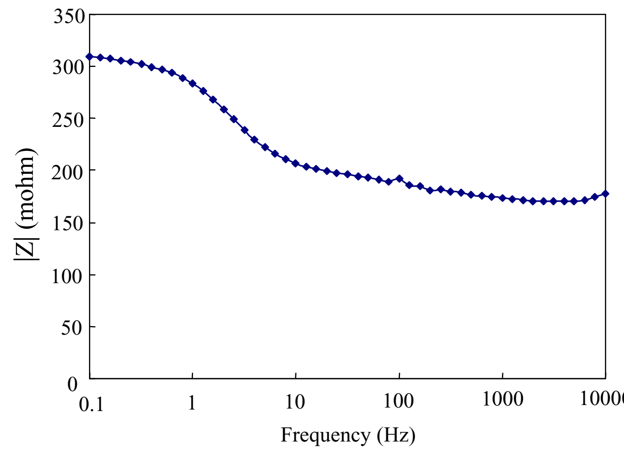

Fig. 9. Bode diagram (|Z|) data for Li-ion battery pack.

Figs. 9 and 10 indicate absolute value $|Z|$ and the phase theta of impedance in Fig. 8. The frequency in Fig. 10, in which the impedance of the imaginary part is zero, is about 1–2 kHz. This impedance is equal to R0. Moreover, as Fig. 9 shows, the value of $|Z|$ is relatively fixed at approximately several kHz. From the above results, it is clear that we can easily obtain a value of R0 by measuring the impedance at about 1 kHz with the simple impedance measuring instrument described above. Here, the absolute value of impedance at 1 kHz is expressed as $|Z(f = 1\,\text{kHz})|$.

2.3. Relationship between capacity deterioration and impedance

Fig. 11 shows three kinds of Cole–Cole plot data, the first for fresh Li-ion battery packs (one cycle) and the other two for deteriorated battery packs (450 and 800 cycles) measured with the same equipment. The relative battery capacity in the first cycle is equal to about 100% (100% is defined as relatively fresh battery capacity), and the relative battery capacity for the 450th and the 800th cycles is 79.8 and 25.6%,

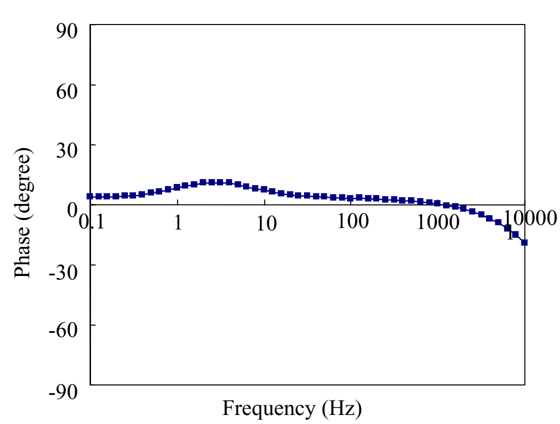

Fig. 10. Bode diagram (phase) data for Li-ion battery pack.

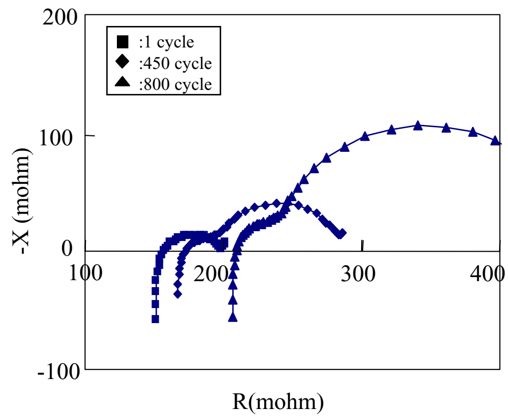

Fig. 11. Cole–Cole plot of deterioration characteristics.

respectively. From the figure, it is obvious that there are two changes in the curves that depend on battery deterioration. The first is the change in impedance (R0), in which the imaginary part is zero. Impedance (R0) increases incrementally in relation to the number of cycles. The second is the change in the two half-cycles in the Cole–Cole plots at a frequency of several 100 Hz. The diameter of the two half-cycles has increased and the center of two half-cycles has moved obliquely to the upper right in relation to the increments in the number of the cycles. We consider that this phenomenon is based on the incrementing of Rp1 and Rp2 in Fig. 7, but it is difficult to evaluate these values precisely because the two half-cycles are not isolated completely.

Fig. 12 shows capacity and absolute value data for impedance at 1 kHz $|Z(f = \text{kHz})|$ for the 1st–800th cycles. It is obvious that the absolute value $|Z(f = 1\,\text{kHz})|$ is nearly R0 according to the results in Figs. 9 and 10. It is also clear from the figure that the absolute value $|Z(f = 1\,\text{kHz})|$ data for impedance at 1 kHz has a strong relationship with battery deterioration.

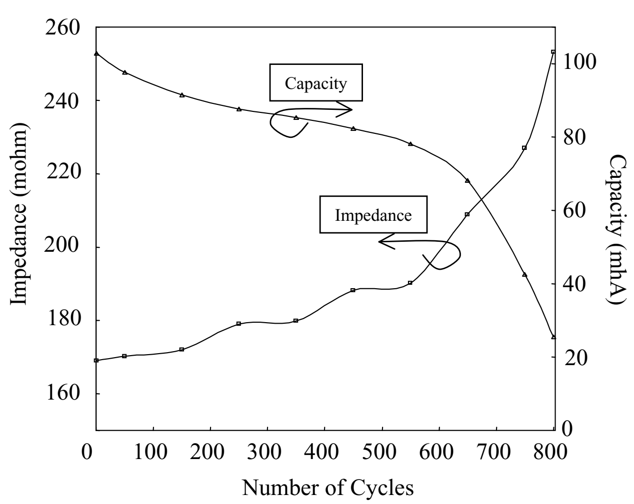

Fig. 12. Deterioration characteristics of battery capacity and impedance (1 kHz).

The cause of the change in impedance R0 is considered to be due to the decrease in the effective surface area of the positive electrode and negative electrode or due to the loose contact between active materials and current collector with the increased number of cycles. The decrease in liquid electrolyte during both electrodes is also considered to cause a decline in the level of electric conduction in the electrolyte and the incrementing of R0 [5,6].

2.4. *Charge condition and impedance of Li-ion battery pack*

The basic and most reliable way of evaluating the deterioration in Li-ion battery packs involves conducting a capacity test where a fully charged battery pack is discharged until the battery voltage drops to the end voltage. However, the main problem with a capacity test is that it takes a long time to complete. An ideal solution would be quick diagnosis without processing regardless of the degree of charge or without having to charge at all before the test.

To achieve this, we investigated how measured impedance changes when states of charges differ and prepared three kinds of battery packs with different levels of charge. These were: (a) a 100% fully charged battery pack that had been charged for 3 h at a constant current of 1 C mA and 4.1 or 4.2 V constant voltage (CC–CV) conditions, (b) a 50% charged battery pack discharged at 1 C mA for 30 min after 3 h of charge, and (c) a 100% discharged battery pack discharged to 3 V (discharge end voltage) after 3 h of charge.

Fig. 13 shows a Cole–Cole plot for Li-ion battery packs with these three states of charge. From the figure, the 100% charged battery pack and the 50% charged battery have almost the same locus. The 100% discharged battery pack has a different locus whose semicircular radius on the lower-frequency side (to the right) reflects a decrease in the reaction velocity at the electrode. However, the portion on the higher-frequency side has the same locus as the other

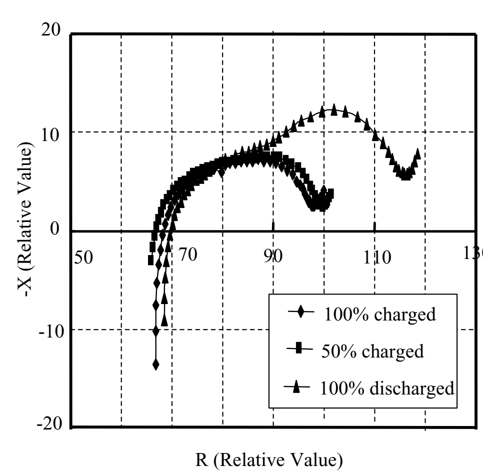

Fig. 13. Cole–Cole plot of deterioration characteristics.

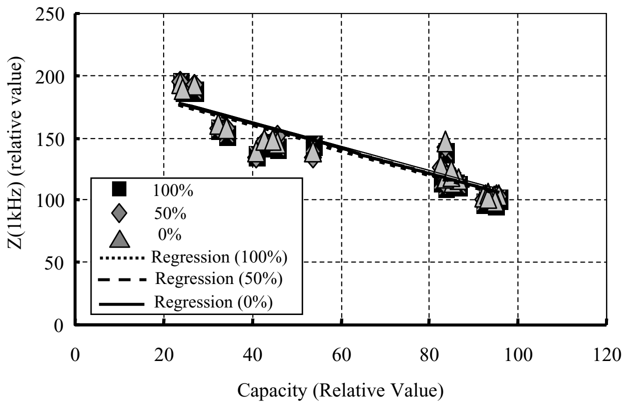

Fig. 14. Z (1 kHz) and capacities in various charge states.

two. This means that neither R0 nor $|Z(f = 1\,\text{kHz})|$ changes much, even if the state of charge changes over a large range (from 100% fully charged to 100% discharged).

Fig. 14 shows the relationship between battery capacity for a fresh battery pack and various degraded kinds and their $|Z(f = 1\,\text{kHz})|$ measured with these different states of charge. $|Z(f = 1\,\text{kHz})|$ has the same value even if the state of charge is different for the same battery pack and this tendency is not related to the degree of deterioration. The lines in the figure are the linear regression of battery capacity and $|Z(f = 1\,\text{kHz})|$ at the three levels of charge, and the squares, diamonds and triangles and the three lines indicate very close agreement.

2.5. Impedance in protection circuit and PTC devices

Two series-FET switches and a PTC device were connected to the Li-ion battery (cell) in-series in the battery pack. The FET is a semiconductor switch and is considered to be less affected by deterioration than Li-ion batteries are by charging/discharging. The PTC device is a reversible electric temperature fuse, which is made from a conductive polymer blend of specially formulated plastics and conductive particles [7]. The PTC device can also be defined as a resistor in an equivalent circuit and may be not influenced by deterioration.

Fig. 15 shows impedance $|Z(f = 1\,\text{kHz})|$ data for a Li-ion battery, a PTC and two-series FETs (on-state) in battery packs with different states of deterioration in battery capacity. It is obvious that impedance $|Z(f = 1\,\text{kHz})|$ data for the PTC and two-series FETs in the battery packs does not change although it does for the Li-ion battery. The value of inductance L, which is based on the wiring in the battery pack circuit, does not change when the wring head does not change. Therefore, the value of L is not influenced by battery deterioration and impedance $|Z(f = 1\,\text{kHz})|$ data.

From Fig. 15, we concluded that the impedance change in a Li-ion battery pack with protective circuit undergoing repeated charge and discharge was similar to that of a battery (cell) and not that of FETs or the PTC.

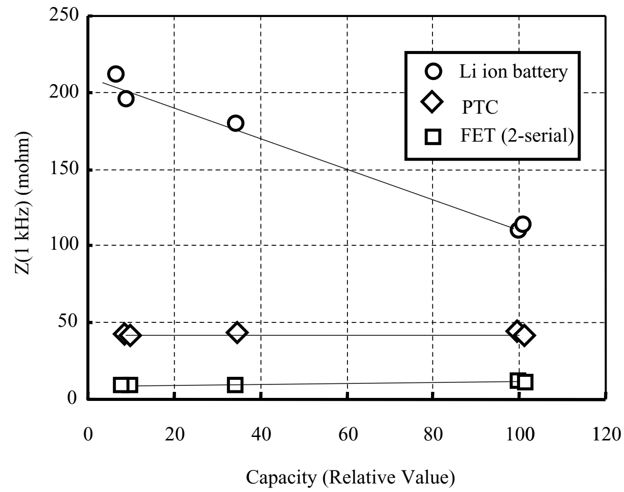

Fig. 15. PTC and FET characteristics of Li-ion battery pack.

2.6. Measurement condition for capacity deterioration

From the above Li-ion battery pack measurements, the following became clear. (a) The absolute value of impedance hardly changes at frequencies of around several kHz (from Fig. 9) and the absolute value of impedance around 1 kHz reflects a value of R0 (from Fig. 10). (b) The impedance of frequencies around several kHz is not greatly influenced by the charging state of the Li-ion battery (from Fig. 13). (c) The impedance of frequencies around 1 kHz has a strong relationship with battery capacity deterioration (from Fig. 14).

The above results indicate that the best test frequency is around 1 kHz because impedance at this level is related to battery capacity deterioration and is not greatly influenced by the charging state. We measured the impedance of battery packs under these conditions using a simple impedance-measuring instrument (frequency fixed at 1 kHz). We confirmed their relationship between impedance (1 kHz) and deteriorating capacity, including accuracy of measurement, using numerous commercially used Li-ion battery packs.

3. Results and discussion

3.1. Regression in estimating battery capacity

We measured $|Z(f = 1\,\text{kHz})|$ and battery capacity in terms of deterioration and clarified the relationships between them. Therefore, if the formula for linear regression is calculated on the basis of results obtained by measuring the impedance of battery packs degraded to various rates of capacity, the battery capacity can be estimated by measuring impedance using a regression formula. It may be the best to derive regression with a logarithm for impedance rather than linear regression since the increase speed of impedance becomes large rapidly as the battery deteriorates. However, we

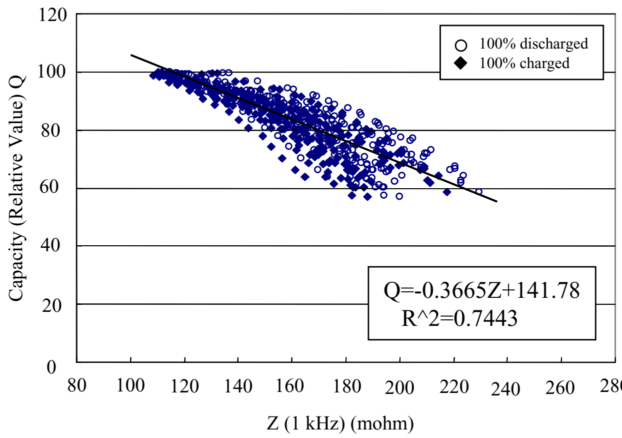

Fig. 16. Linear regression for battery pack type A.

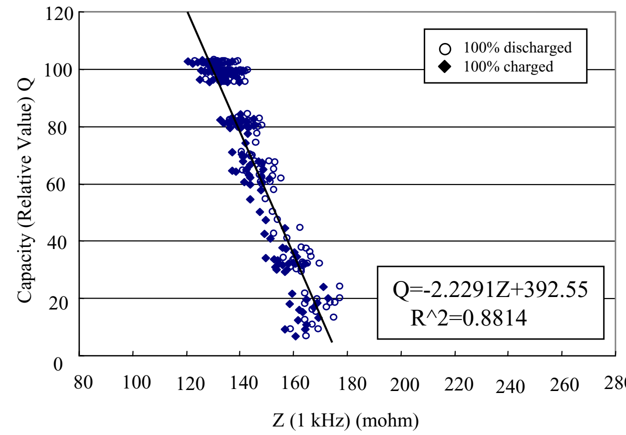

Fig. 19. Linear regression for battery pack type D.

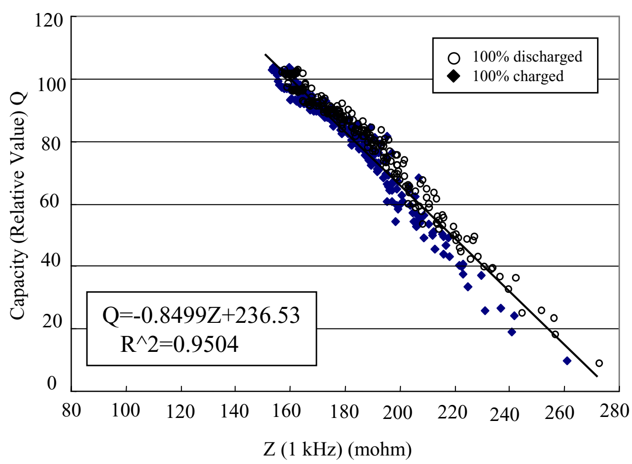

Fig. 17. Linear regression for battery pack type B.

selected linear regression because it is practical and simple to calculate.

Figs. 16–19 are respective linear regressions for battery pack types A–D, which have different manufacturers. Table 1 shows the specifications and measurement data

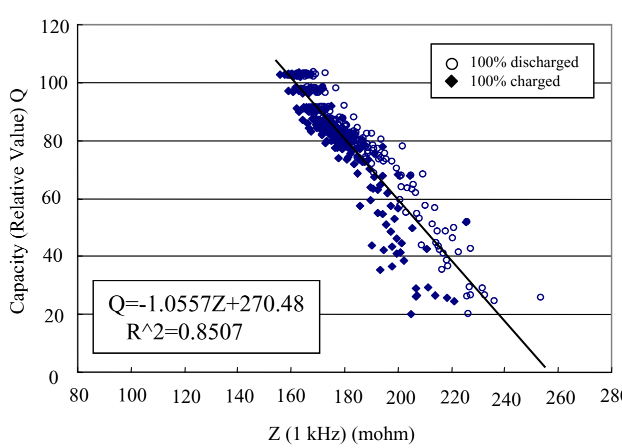

Fig. 18. Linear regression for battery pack type C.

of these battery packs. Although the plotted data was distributed around the linear regressions, on the whole, there seemed to be a high correlation coefficient between $|Z(f = 1\,\text{kHz})|$ and battery capacity. Therefore, we expect that the extent of deterioration of a battery can be assessed easily and quickly by using this formula for linear regression and by measuring the impedance of the battery. In Figs. 16–19, the slopes for linear regressions vary by battery model manufacturer.

The product of a battery's capacity value as multiplied by its impedance value in Table 1 is considered to be ideally constant because the battery capacity is proportional to the battery–electrode area. This value is also inversely proportional with its battery impedance when these battery packs are almost equal, having similar internal structure, similar material, and nearly the same energy densities (about 155 Wh/kg). From Table 1, it is clear that the product values of the battery capacity by its impedance are nearly constant, from 61,880 to 67,600. Assuming that the battery's electrochemical characteristics are desirable when its slope value of linear regression is as high as possible, the characteristics of type D is best, types C and B perform less well, and type A the least desirable of the four types of battery packs.

About the difference of slope values at linear regression in Table 1, the slope value is indicated as the change rate of impedance against the change of the degraded capacity. It is thought that the battery electrochemical characteristics are as good because the slope value is larger (the change of impedance is lower against the change of capacity). From Table 1, the slope value of type D is the largest (best), and it is the order of type C, B, and A. Though these battery characteristics are almost equal, the only main difference of these batteries is the electrolyte layer. The battery of type A has a polymer material separator in the electrolyte layer and battery types B, C, and D have a polyethylene material separator. It is thought that the impedance sensitivity (rate of change) against the change of capacity with the polymer material separator is higher than that with the polyethylene material separator by the influence in the conductivity

Table 1
Specifications of measured battery packs

Type	Absolute value Q0 (mho A)	Impedance (mΩ) $\|Z(f = 1\,kHz)\|$				Q0 × Z (Z = Za − R1)	Slope value of linear regression
		Za (Q = 100%)	Zb (Q = 50%)	Rate of Zb/Za	R1		
A	650	114	250	2.2	10	67600	−0.37
B	600	160	219	1.4	50	66000	−0.85
C	570	161	209	1.3	50	63270	−1.06
D	680	131	154	1.2	40	61880	−2.23

characteristics (polymer separator is lower than polyethylene one), and the contained quantity of electrolyte (polymer separator is lower than polyethylene one). Regarding the differences between types B, C, and D, the possibility of mixing additional materials to the electrolyte (which is mainly the organic solvent of the lithium salts) exists because the main materials and internal structures being used for these batteries are almost same.

From the above explanation, it has been suggested that it is necessary to calculate the regression formula for each model because each model has a different design concept.

Fig. 20 presents errors in estimating the battery capacity of about 800 battery packs using impedance $|Z(f = 1\,kHz)|$ and a linear regression formula calculated for each battery pack model. About 70% of the total results have estimation errors of less than ±10%, and if the error range is extended to less than ±20%, 90%, or more of the data is included. As a result, we expect that this method will provide quick and simple deterioration diagnosis that is very efficient.

3.2. Standard to assess deterioration

We cannot generally assess to what extent a degraded battery pack has been used, i.e., whether it has been used until capacity deteriorates to whatever percent because this depends on system design and how it has been used. However, in our evaluation, we assumed that they had been used until they deteriorated to about 50% of nominal capacity, and established a new standard with three stages of degradation (Table 2) to simplify the process.

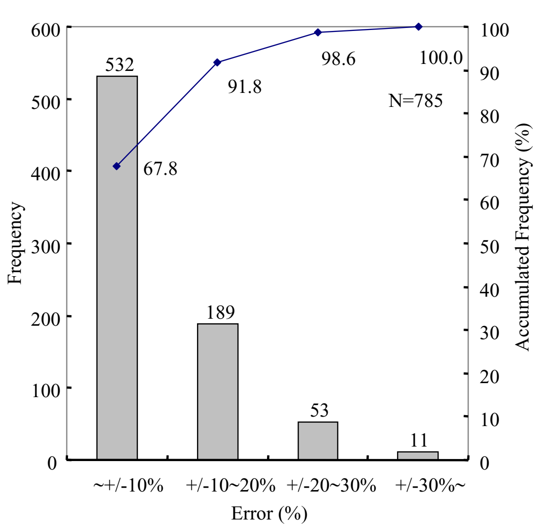

Fig. 20. Accuracy of estimating capacities (all data).

Table 2
Standard to assess deterioration in battery packs

	Degree of battery capacity	Estimated capacity
1	Good	60% or more
2	Fair	60–40%
3	Needs to be exchanged	40% or less

We tested about 800 commercial battery packs, some were new, and the others were mainly degraded through repeated discharging/charging cycles. We correctly classified them into three groups on the basis of our new standard and capacity, estimated from the results of measuring impedance $|Z(f = 1\,kHz)|$, using the regression formula. These results are plotted in Figs. 21–23 with all the data for 100% charged, 50% charged, and 100% discharged battery packs.

Fig. 21 shows that about 95% of battery packs assessed to be "good" actually had 60% or more capacity. Fig. 22 has assessments for "fair" (capacity between 40 and 60%), which was the worst of the three classified groups. Battery packs correctly classified into this group represented about 55% of the whole. About 35% of battery packs classified into this group only had 40% or less capacity while about 5% had 60% or more capacity. Batteries that "needed to be changed" (capacity of less than 40% in Fig. 23) represented about 80% of battery packs. Several battery packs classified into this group actually had a capacity of 80% or more.

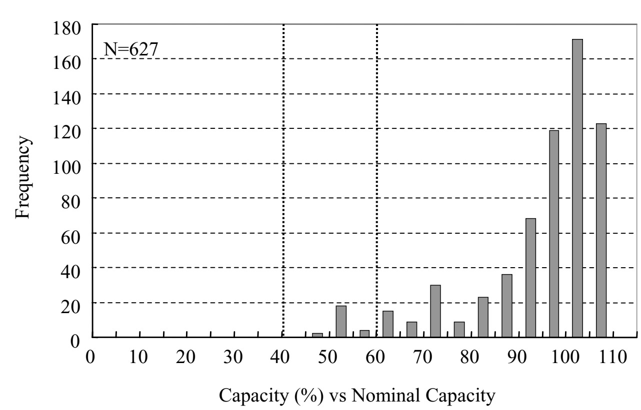

Fig. 21. Capacity distribution for batteries judged to be "good" by quick deterioration diagnosis.

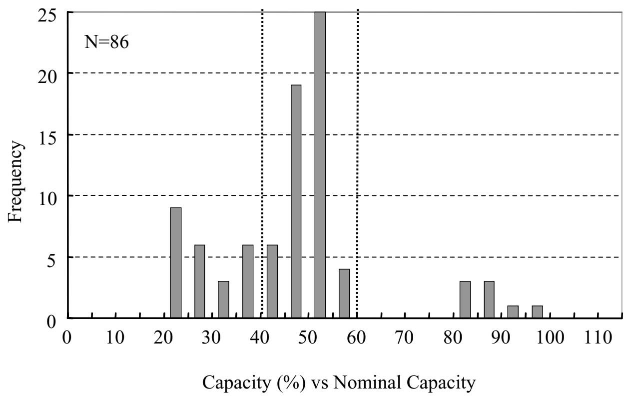

Fig. 22. Capacity distribution for batteries judged "fair" by quick deterioration diagnosis.

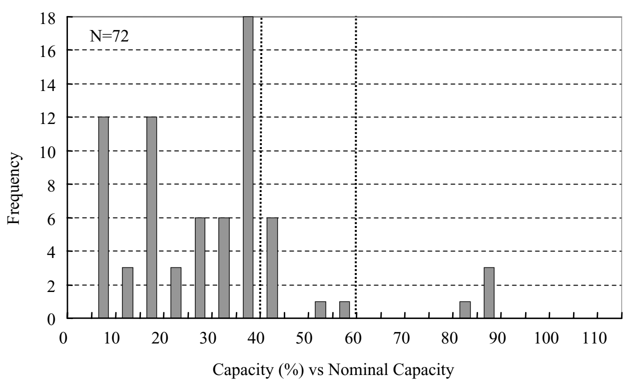

Fig. 23. Capacity distribution for batteries that were judged as "need to be changed" by quick deterioration diagnosis.

As a whole, the capacities of about 90% of all batteries were judged correctly. This method involves measuring impedance at 1 kHz and it shows promise as a quick and simple means of assessing deterioration in Li-ion batteries.

4. Conclusions

We measured numerous impedance data for various fresh-to-degraded Li-ion battery packs and reached the following conclusions.

In the experiment, we found that the best test frequency was around 1 kHz because impedance at this frequency is related to deteriorating battery capacity and is not greatly influenced by the degree of charge. We did tests on about 800 commercially available Li-ion battery packs for mobile phones, which we degraded by subjecting them to repeated cycles of discharge/charge. This resulted in battery packs with a variety of capacity levels. We then investigated the relationship between impedance measurement results at 1 kHz and remaining capacity in the battery packs. Consequently, we found that they were closely related. We calculated a formula for linear regression to estimate the capacity of each battery pack model, and found that the capacities estimated from impedance using the formula had errors of no more than 20% for about 90% of the 800 battery packs. Finally, we did quick and simple three-stage assessments and correctly evaluated approximately the capacities of 90% of all battery packs regardless of their state of charge.

In this paper, we described quick diagnosis that is mainly intended for the evaluation of battery packs that have been degraded through repeated discharge and charge cycles. However, the results did not reflect the actual state of battery packs in some cases. We need further studies on deterioration modes other than cyclic discharging and charging, such as overcharging, overdischarging, long-term storage, and exposure to high-temperatures.

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