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Novel State-of-Charge Estimation Method for Lithium Polymer Batteries Using Electrochemical Impedance Spectroscopy

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Abstract

Lithium batteries are widely used in mobile electronic devices due to their higher voltage and energy density, lighter weight and longer life cycle when compared to other secondary batteries. In particular, a high demand for lithium batteries is expected for electric cars. In the case of the lithium batteries used in electric cars, driving distance must be calculated accurately and discharging should not be done below a level that makes it impossible to crank. Therefore, accurate information on the state-of-charge (SOC) becomes an essential element for reliable driving. In this paper, a novel method for estimating the SOC of lithium polymer batteries using AC impedance is proposed. In the proposed method, the parameters are extracted by fitting the measured impedance spectrum on an equivalent impedance model and the variation in the parameter values at each SOC is used to estimate the SOC. Also to shorten the long length of time required for the measurement of the impedance spectrum, a novel method is proposed that can extract the equivalent impedance model parameters of lithium polymer batteries with the impedance measured at only two specific frequencies. Experiments are conducted on lithium polymer batteries, with similar capacities, made by different manufacturers to prove the validity of the proposed method.

Key Words: Electrochemical Impedance Spectroscopy, Lithium Polymer Battery, Randle's Circuit, State-of-Charge, Time Constant

I. Introduction

With the increasing complication of mobile electronic devices, the need for high performance, highly safe, environmentally friendly, super light energy storage devices is also increasing. Lithium-ion batteries are receiving a great deal of attention as secondary batteries that satisfy these conditions. Lithium-ion batteries have higher than average voltage and energy density, a lower self-discharge, a longer life and no memory effect when compared to Ni-MH and Ni-Cd batteries [1]-[3]. However, it is well known that lithium-ion batteries also have a risk of explosion, a high manufacturing cost and difficulty in enlargement. As a means to resolve such problems, a lithium polymer battery has been proposed. A lithium polymer battery is composed of positive and negative electrodes where oxidation and reduction occur, a separation membrane which prevents physical contact between the two electrodes, and a solid or gel-type electrolyte with the role of a medium for ion movement.

When a lithium polymer battery is used for mobile purposes, the running time of the electronic device being used must be known. If the battery is supplying power to an important facility or processing network, sudden shutdowns from a discharge should be avoided. Also, in case of large capacity lithium polymer batteries for electric cars, accurate calculation of the traveling distance is required to guarantee that the battery is not discharged in a way that makes cranking impossible.

Since the state-of-charge (SOC) of a battery cannot be measured directly, it is often measured by an indirect method using either the terminal voltage or the current. The residual capacity (or SOC) of a battery can rarely be accurately determined using only the terminal voltage due to its non-linearity. In particular, there are not many established methods for estimating the SOC of lithium polymer batteries. The method most often used for the estimation of SOC in secondary batteries is Coulomb counting, which calculates the amount of charge by counting the charge / discharge current of the battery. Although extremely accurate and reliable information can be obtained with an accurate current sensor, it is difficult to apply this method to dynamic systems in which large charge / discharge current values alternate in a very short period of time because of error accumulation [4].

A Kalman filter can be used to estimate the SOC of a battery, but the algorithm is complex and accurate system modeling is essential. For accurate estimation, there is the problem of too many state variables that must be considered. In addition, due to the complicated gain configuration system the gain may diverge if there is a noise or disturbance added to the system. One of basic presumptions of a Kalman filter is that disturbances or process noise must have a random Gaussian distribution. However, this presumption is seldom

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met in actual operating conditions, so Kalman filters cannot always be operated in an optimum state [5]-[7].

Another method is to use electrochemical impedance spectroscopy using the parameters of an equivalent impedance model. This method measures the AC impedance spectrum for each SOC of a battery through electrochemical impedance spectroscopy and fits it using an equivalent impedance model. The parameters of the model are extracted and the SOC of the battery is estimated by analyzing the correlation between the parameters and the SOC [8]-[12].

There are other methods like neural networks [13]-[14] and fuzzy logic [15]-[16], but the former requires highly empirical components in its design and there is no guarantee that the estimation error converges to the global minimum. This requires repetition of the method with different initial values. The latter requires a complicated system abstracted from knowledge and experience. Therefore, these two methods are rarely used in actual systems due to the fact that they require too many calculations and have limited reliability. This paper proposed a new method for estimating the SOC of lithium polymer batteries using electrochemical impedance spectroscopy. Specifically, this method extracts the parameters of an equivalent impedance model by using impedance values measured at two frequencies. The usefulness of the proposed method has been verified through experiments on lithium polymer batteries with similar capacities made by different manufacturers.

II. Experimental

A. Decision of Rated Capacity through Charge and Discharge

The SOC of a battery refers to the amount of charge which can be released from that battery at a specific point, expressed against the rated charge as shown by Eq. (1).

$$SOC = \frac{Q_{releasable}}{Q_{rated}} \times 100\%. \tag{1}$$

First, charge/discharge experiments were conducted to check the rated capacity of the battery. After charging the battery in current control mode with a current of 1C up to a maximum of 4.2V, the mode was changed to voltage control mode with a voltage of 4.2V. The battery was charged again until the current reached almost zero (20mA). This is called the CC-CV charging method. Then in accordance with the IEC 61960 regulation, the amount of charge in the lithium polymer battery was measured while discharging under a constant current of 0.2C from a maximum voltage of 4.2V to a cut-off voltage of 3.0V. The waiting time between charging and discharging was chosen to be two hours for the experiment, and charging was performed up to five times if the battery failed to reach its rated capacity. In this case, the maximum value among the five was selected to be the rated capacity [17]. The battery used was a commercial lithium polymer battery manufactured by BNK with a normal voltage, a maximum voltage, a cut-off voltage and a normal capacity of 3.7V, 4.2V, 3V and 1600mAh, respectively.

Fig. 1 is a curve showing the relationship between the charge current and the battery voltage obtained in the experiment on the BNK lithium polymer battery. Fig. 2 shows a voltage curve obtained from the experiment with a constant current of 0.2C. The SOC of the lithium polymer battery can be calculated according to the discharge time based on the rated capacity (1568mAh) found in the charge-discharge experiment.

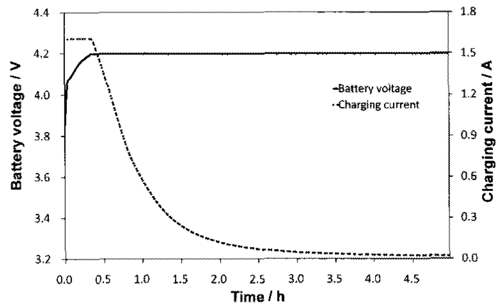

Fig. 1. The charging curve of the BNK lithium polymer battery.

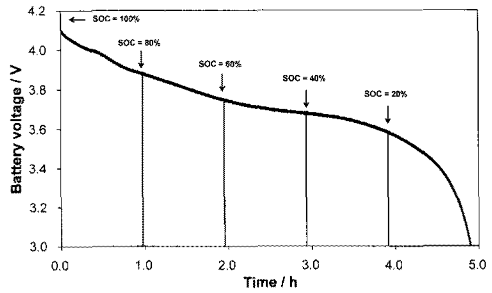

Fig. 2. The discharging curve of the BNK lithium polymer battery.

B. Electrochemical Impedance Spectroscopy

Electrochemical impedance spectroscopy (EIS) induces a small perturbation near the target, measures the AC impedance from the response to the perturbation, fits the curve using an equivalent impedance model that can physically explain the measured AC impedance, and models the target [6]. Since impedance reflects the internal state as it is, accurate information about the SOC can be obtained by measuring the impedance and extracting the parameters of an equivalent circuit with EIS.

A BPS 1000FL was used as an experimental instrument for obtaining the AC impedance spectrum of the battery, and the measurement of the impedance was performed at frequencies between 10mHz~1kHz. The perturbation current was chosen to not exceed 5% of the charge variation in the battery in order to guarantee the linearity of the experiment, the response voltage by perturbation was kept below 50mV, and the DC bias was 0mV. The measurement of the impedance was performed at room temperature (25°C) for the range of a 20~100% charge state with a 20% interval. The battery was left intact for two hours between each step, and AC impedance was measured through EIS.

III. Results and Discussion

A. Impedance spectrum of the lithium-polymer battery

Fig. 3 shows the impedance spectrum of the BNK battery measured at each SOC by EIS. The impedance spectrum

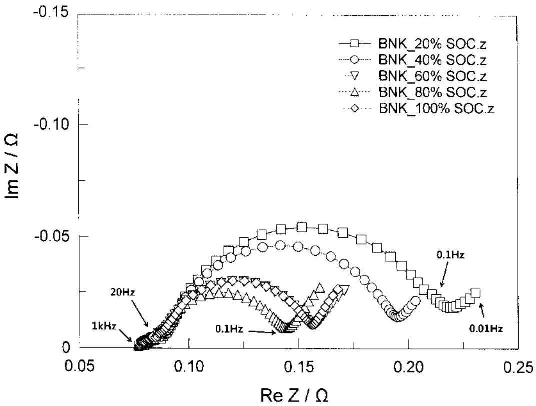

Fig. 3. Impedance spectra of the BNK lithium polymer battery at each SOC.

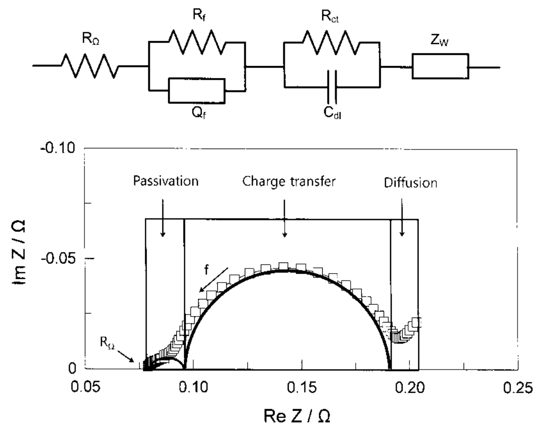

Fig. 4. Impedance spectrum and equivalent circuit of the lithium battery.

crosses the real axis at Re $Z \approx 0.75\text{m}\Omega$ and a large semi-circle exists in the frequency range of 20Hz~0.1Hz. Also, in the low frequency region below 0.1Hz, the Warburg impedance is spreading out at an angle of 45° from the real axis. Looking at the impedance for each SOC, a decrease in the SOC of a lithium battery does not change the shape of the impedance spectrum in the frequency range of 1kHz~20Hz. In contrast, the large semi-circle experiences an increase in radius. This is a phenomenon that results from the increase in the internal impedance due to the discharging of the battery. Accordingly, the SOC can be estimated by analyzing the parameters of an equivalent impedance circuit that shows the semi-circle in the low frequency region.

B. Chemical Reactions of a Lithium Polymer Battery and an Impedance Equivalent Circuit

Representative chemical reactions of lithium polymer batteries such as passivation, charge transfer and diffusion reactions have different time constants. Therefore, they are observed at different frequencies on a Nyquist impedance plot. Fig. 4 represents the impedance plot of a BNK lithium polymer battery measured at an SOC of 40%. There are many types of equivalent circuits for lithium polymer batteries, but a circuit with R_Ω, ZARC(R_f//Q_f), R_{ct}//C_{dl} and Z_W, representing ohmic resistance, passivation, charge transfer and diffusion, connected in-series, was used to analyze the impedance spectrum of a lithium polymer battery (Fig. 4).

TABLE I
IMPEDANCE PARAMETERS OF THE BNK LITHIUM POLYMER BATTERY

	SOC (%)				
	20	40	60	80	100
R_{ct} (mΩ)	93.5	80.	50.0	40.7	52.6
C_{dl} (F)	2.74	2.69	2.65	2.67	2.48
$\tau = R_{ct} \times C_{dl}$	0.27	0.22	0.13	0.11	0.13

In an equivalent circuit, the ohmic resistance R_Ω refers to the overall resistance that connects the electrodes of a battery with the inside, namely the electrolyte, the electrode and the terminal block. This is the value given at the point where the curve crosses the real axis on the Nyquist plane. In lithium batteries with the passivation phenomenon, a small semi-circle in the impedance spectrum is shown in the high frequency region. This can be modeled as a parallel structure of a resistance and a capacitor. However, as seen in Fig. 4, the small semi-circle in the high frequency region is slightly depressed and can be more accurately expressed as a ZARC, a parallel circuit with resistance (R_f) and a constant phase element (CPE : Q_f) [18]-[19].

This small semi-circle shows the components of a passivation film created on the surface of the negative electrode when the battery is used. Since the thickness of the film is not consistent, it can only be expressed properly using a CPE as shown by Eq. (2) [19]-[21].

$$Z_{CPE} = \frac{1}{Q(j\omega)^n}. \tag{2}$$

A large semi-circle which shows the charge transfer reaction is expressed as a parallel connection of the charge transfer resistance (R_{ct}) with a double layer capacitance (C_{dl}). The large semi-circle shows an electron transfer reaction between two electrodes (positive and negative) and the electrolyte [19], [22].

The fitting of curves with the equivalent circuit of Fig. 4 was done using Zview. As explained earlier, the parameters related to the large semi-circle, for which there are clear differences in the impedance plot according to the different SOCs, are summarized in Table I. The trend of the parameters for each SOC is shown in Fig. 5. Looking at R_{ct} and C_{dl} which are the parameters of the R-C circuit related to the large semi-circle, R_{ct} shown as the radius of the large semi-circle, decreases in the SOCs between 20%~80% and increases in the SOCs between 80%~100%. This result corresponds to the size change in the semi-circles of a Nyquist impedance plot. On the other hand, C_{dl} shows a change in the parameter value according to a change in the SOC. However, this change is less than 10%. Also, the best-fit line for parameter variations according to the SOC was deduced through a numerical analysis and the relationship between them was explored. As illustrated in the figure 5, the SOC can be estimated exclusively using R_{ct}. An accurate estimation of the SOC is also possible using a time constant, which is a product of R_{ct} and C_{dl}.

IV. EXPERIMENTS WITH BATTERIES BY DIFFERENT MANUFACTURERS

In case of a BNK battery, the SOC was successfully estimated using the change in the R_{ct} parameter or the time

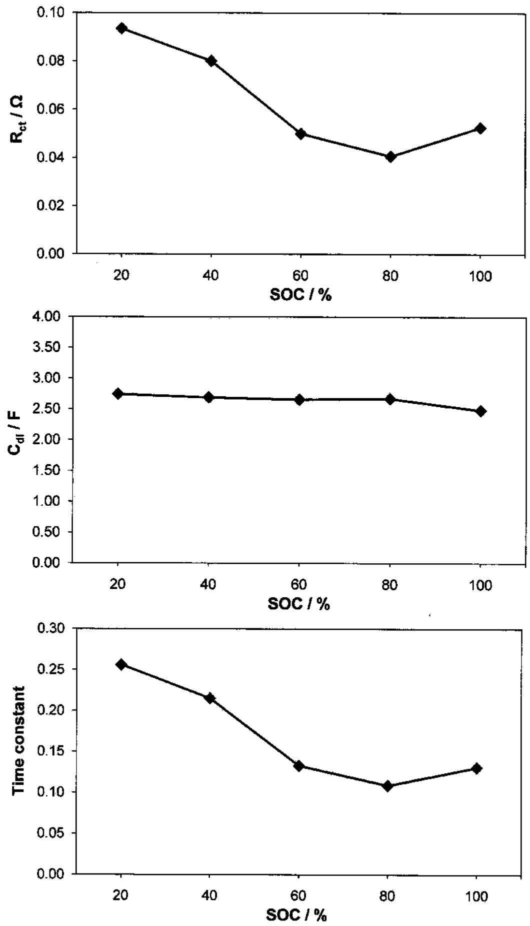

Fig. 5. Parameters of the BNK lithium polymer battery at each SOC (a) R_{ct} (b) C_{dl} (c) Time constant.

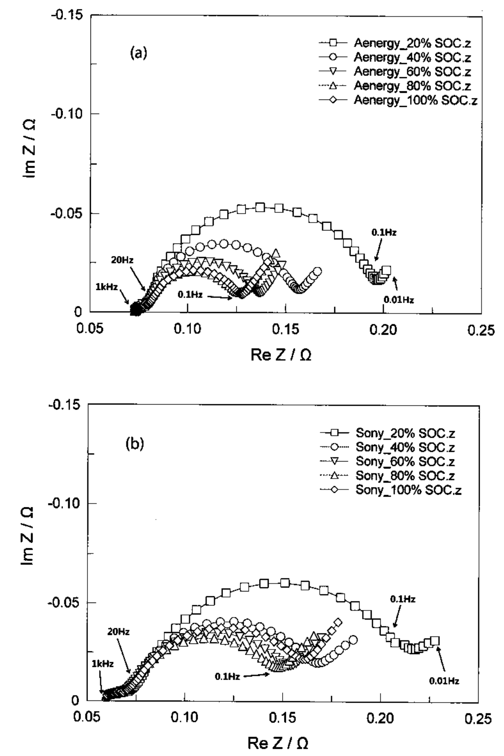

Fig. 6. Impedance spectra of the batteries at each SOC (a) Aenergy (b) SONY.

TABLE II
IMPEDANCE PARAMETERS OF THE AENERGY LITHIUM POLYMER BATTERY

	SOC (%)				
	20	40	60	80	100
R_{ct} (mΩ)	96.4	63.4	46.7	36.4	37.5
C_{dl} (F)	3.27	3.26	3.23	3.42	3.41
$\tau = R_{ct} \times C_{dl}$	0.31	0.21	0.15	0.12	0.13

TABLE III
IMPEDANCE PARAMETERS OF THE SONY LITHIUM POLYMER BATTERY

	SOC (%)				
	20	40	60	80	100
R_{ct} (mΩ)	105	63.1	55.7	51.3	58.1
C_{dl} (F)	4.40	4.47	4.19	4.20	4.57
$\tau = R_{ct} \times C_{dl}$	0.46	0.28	0.23	0.22	0.27

constant according to the changes in the SOC. Here, an experiment was conducted to verify that the SOC could be estimated using R_{ct} or the time constant for all lithium polymer batteries regardless of manufacturer.

Commercial lithium polymer batteries with similar capacities manufactured by Aenergy, Sony and BNK were selected as experimental subjects. The nominal voltage, maximum voltage and cut-off voltage of these batteries are identical to those of the BNK battery above at 3.7V, 4.2V and 3V, respectively. The nominal capacities are 1800mAh and 1350mAh. In order to determine the rated capacity of the batteries from the two other manufacturers, the charge-discharge experiment described in the previous section was performed. Based on this, the SOC of the lithium polymer batteries was calculated according to the discharge time. The impedance spectrum measured with EIS was curve-fitted using the equivalent circuit in Fig. 4 and the parameters of the equivalent circuit were extracted. The impedance spectrum of the Aenergy and Sony batteries are shown in Fig. 6. The parameters of the models extracted through fitting are given in Table II and Table III.

Looking at the impedance spectra of the Aenergy and Sony batteries in Fig. 6, a small semi-circle appears in the frequency range of 1kHz∼20 Hz and a large semi-circle appears in the range of 20Hz∼0.1 Hz on a Nyquist impedance plot with shapes similar to those of the BNK battery. The two batteries also showed little change in the size of the small semi-circle in the impedance plot with the progress of discharge. The radii of the large semi-circles of the two batteries are also increased with the progress of discharge. The Warburg impedance can also be found at frequencies below 0.1Hz, which is very similar to what was shown by the BNK battery.

The relationship between the R_{ct} parameter, the time constant and the SOC of the Aenergy, Sony and BNK batteries, extracted from an equivalent impedance model through curve-fitting with Zview, is shown as a graph in Fig. 7. As a result of analysis, it can be seen that the batteries from all three manufacturers show similar trends in the relationship between the R_{ct} parameter and the SOC. The SOC can also be estimated using a time constant, which is a product of R_{ct} and C_{dl}. However, as seen in the figure, the change in parameters for each SOC becomes clearer using a time constant in addition to R_{ct}, which also increases the accuracy of estimation.

Using the parameters of an equivalent circuit, the changes in R_{ct} and a time constant according to the SOC were approximated as a polynomial function of degree 3. The

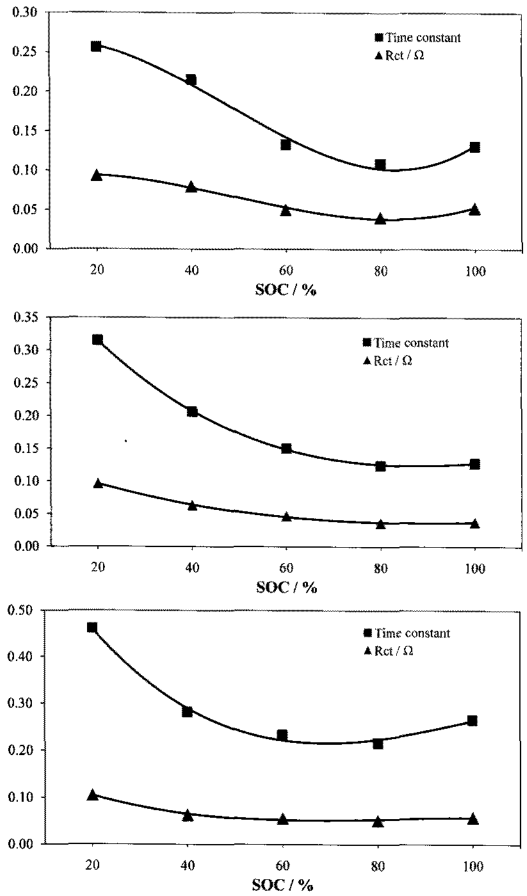

Fig. 7. Parameters of the lithium polymer battery at each SOC (a) BNK (b) Aenergy (c) SONY.

TABLE IV
COEFFICIENT AND FITTING ACCURACY(R2) OF TREND LINE EQUATION

	BNK		Aenergy		SONY	
	R_{ct}	τ	R_{ct}	τ	R_{ct}	τ
a (10^{-7})	3.927	9.135	-0.496	-2.385	-2.431	-6.656
b (10^{-4})	-0.579	-1.315	0.222	0.881	0.617	2.073
c (10^{-2})	0.157	2.884	-0.279	-0.995	-0.497	-1.921
d	0.083	0.245	0.144	0.480	0.181	0.764
R^2	0.990	0.989	0.999	1.000	0.989	0.993

accuracy of the best-fit line on the changes in R_{ct} and the time constant according to the SOC of each battery calculated by the least squares method was over 98%. Therefore, the SOC of lithium polymer batteries can be accurately estimated using the equation of this best-fit line. The best-fit equation against the changes in R_{ct} and a time constant can be expressed as Eq. (3), and the coefficients and accuracy of the fitting are shown in Table IV.

$$\tau(SOC) = a(SOC)^3 + b(SOC)^2 + c(SOC) + d. \tag{3}$$

Accordingly, by performing an EIS experiment on a specific lithium battery and replacing the left term of Eq. (3) with a time constant, the SOC can be estimated by solving a polynomial function of degree 3 on the SOC.

However, Eq. (3) is a polynomial function of degree 3 and three solutions are found by solving the equation by substituting a time constant into the left term. In general, one feasible and two infeasible solutions are found.

Looking at Fig. 7 in detail, there is a section in which two

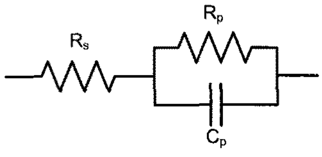

Fig. 8. Randle's circuit.

feasible solutions are found. For example, in case of a BNK battery, as shown by Fig. 7 (a), if the solutions are found using Eq. (3) and a time constant measured at around 80% SOC, the parameter between 60%~80% and the parameter between 80%~100% take on symmetric values, resulting in two SOC values. This problem can be solved by using the slope of continuously estimated SOCs. The slope of the change in the SOC can be calculated by using two consecutive SOC estimations and equation (4).

$$\frac{\Delta\tau}{\Delta SOC} = \frac{\tau_{old} - \tau_{new}}{SOC_{old} - SOC_{new}}. \tag{4}$$

Then, two sets of estimated SOC values are obtained, one with a positive value and the other with a negative value. If the battery is in the charging process, then the one with the negative value is the correct estimation and if the battery is in the discharging process, then the one with the positive value is the correct estimation.

V. SIMPLIFIED METHOD FOR THE EXTRACTION OF EQUIVALENT CIRCUIT PARAMETERS

Using the proposed method, the SOC of lithium batteries was found with a time constant from EIS. However, EIS measures the impedance spectrum for each frequency and calculates the parameters of an equivalent impedance circuit based on EIS results. Since it takes a long time to conduct an experiment for a target frequency range of 10mHz~1kHz, it is difficult to make use of this method in actual applications. Therefore, this paper proposes a novel method that can actually be applied to commercial applications. The new method estimates the SOC using the impedance measured at two specific frequencies using the geometric characteristics of a Nyquist impedance plot. First, a Nyquist impedance plot of a lithium polymer battery can briefly be expressed as a Randle's circuit as shown in Fig. 8. As explained earlier, the ZARC impedance in the high frequency region by passivation shows no change with changes in the SOC. Since a change in the impedance from changing the SOC mainly appears in the large impedance semi-circle in the low frequency region, there is no problem in simplifying lithium battery impedance into a Randle's circuit. As shown in Fig. 9, the central coordinates $(x_1, 0)$ of the semi-circle on the real axis and the radius of the semi-circle can be found using a system of equations based on Eq. (5), if impedance values at two specific frequencies in a Nyquist impedance plot are known. Here, the two specific frequencies must be an appropriate distance away from the cut-off frequency of the semi-circle for accurate calculations. By comparing the impedance plots of the three lithium batteries shown in Fig. 9, all three batteries are shown to be in the frequency range of 1.5Hz~24.5Hz for the impedance that appeared in the left semi-circle. The range was

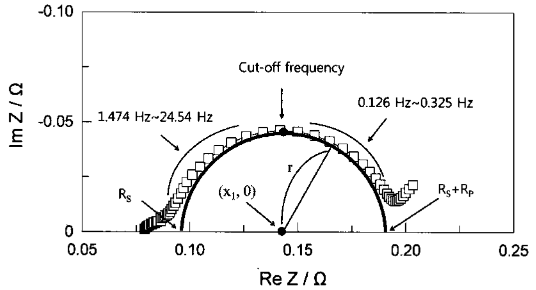

Fig. 9. Frequency characteristic of Randle's circuit and its geometrical characteristics of impedance spectrum.

0.126~0.325Hz for the right semi-circle. Therefore, 1.45Hz and 0.32Hz from the middle of each region were selected as sample frequencies. The two impedances measured at sample frequencies are expressed as (Re Z_1, Im Z_1) and (Re Z_2, Im Z_2). The large semi-circle in the low frequency region of a Randle's circuit impedance spectrum can be expressed as an equation for a circle with a center $(x_1, 0)$ and a radius r. By solving the system of equations through the substitution of real and imaginary impedance values measured at two frequencies into Eq. (5), x_1 and r can be found.

$$(\mathrm{Re}\, Z - x_1)^2 + \mathrm{Im}\, Z^2 = r^2. \tag{5}$$

The parameters of R_s and R_p in a Randle's circuit are found by substituting the values of x_1 and r from Eq. (5) into Eq. (6).

$$R_s = x_1 - r, \quad R_p = 2r. \tag{6}$$

Impedance of a Randle's circuit can be shown as Eq. (6), and the value of C_p can be calculated using Eq. (7) by substituting R_s, R_p, the frequency and the impedance.

$$Z(j\omega) = R_s + \frac{R_p}{1 + j\omega R_p C_p}. \tag{7}$$

In order to demonstrate the accuracy of the proposed method, a time constant extracted from a Nyquist impedance spectrum for all frequency ranges and a time constant found using the values of R_p and C_p extracted by the impedance from two frequencies were compared in Fig. 10. An analysis with the least squares method showed an accuracy of 98.11% for a BNK battery, 99.15% for an Aenergy battery and 95.77% for a Sony battery. The proposed method can simply and accurately estimate the SOC of lithium batteries.

VI. Conclusions

A new method for estimating the SOC of lithium polymer batteries was proposed. Lithium polymer batteries are expected to be widely used in the future for mobile electronic devices, hybrid cars and purely electric cars. The SOC of lithium polymer batteries was estimated by the proposed method using changes in the parameters of an equivalent impedance model. This method was verified with excellent accuracy in the estimation of SOC. In particular, the parameters of an equivalent circuit were extracted using the impedance measured at two frequencies using the geometric characteristics of the impedance spectrum of lithium polymer batteries simplified as

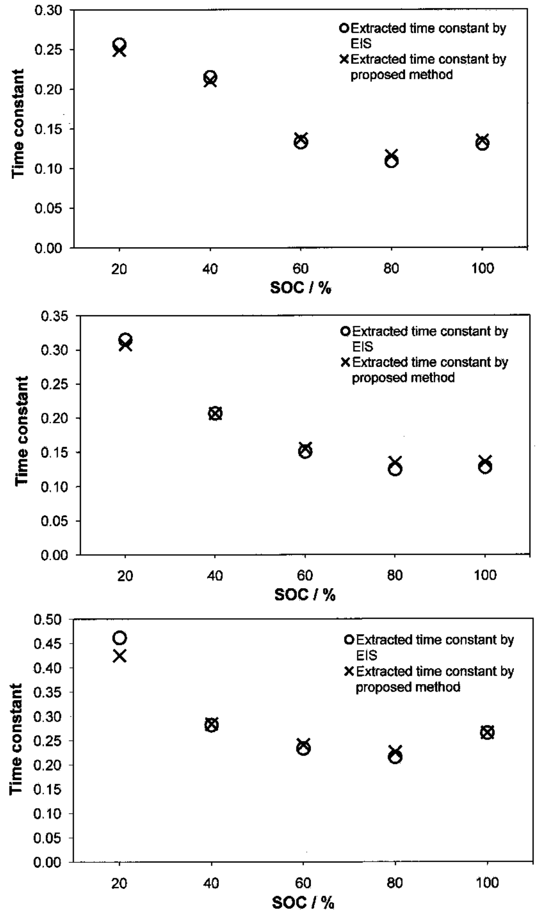

Fig. 10. Comparison of extracted time constant by EIS experiment and extracted time constant by proposed method (a) BNK (b) Aenergy (c) SONY.

a Randle's circuit. Such parameters were applied to three types of batteries made by different manufacturers to demonstrate that SOC can be estimated with over 98% accuracy. Since the proposed method only requires impedance measured at two frequencies, the time required for measurement is shortened by a large amount. Due to its simplicity, the proposed method is expected to see wide application in measuring the impedance spectrum of batteries, fuel cells and any kind of electrochemical device having equivalent impedances expressed as a Randle's circuit.

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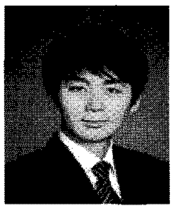

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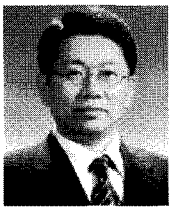

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