ARTICLE

New Li-ion Battery Evaluation Research Based on Thermal Property and Heat Generation Behavior of Battery

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We do a new Li-ion battery evaluation research on the effects of cell resistance and polarization on the energy loss in batteries based on thermal property and heat generation behavior of battery. Series of 18650 cells with different capacities and electrode materials are evaluated by measuring input and output energy which change with charge-discharge time and current. Based on the results of these tests, we build a model of energy loss in cells' charge-discharge process, which include Joule heat and polarization heat impact factors. It was reported that Joule heat was caused by cell resistance, which included DC-resistance and reaction resistance, and reaction resistance could not be easily obtained through routine test method. Using this new method, we can get the total resistance R and the polarization parameter η. The relationship between R, η, and temperature is also investigated in order to build a general model for series of different Li-ion batteries, and the research can be used in the performance evaluation, state of charge prediction and the measuring of consistency of the batteries.

Key words: Li-ion battery, Energy loss, Heat, Cell resistance, Polarization, State of charge prediction

I. INTRODUCTION

Li-ion batteries have been widely used in variety of consumer electronic applications since 1991, because of their high energy density and other advantages. Along with the development of pure electric vehicles (EVs), hybrid electric vehicles (HEVs) and plug-in hybrid vehicles (PHEVs), Li-ion batteries have a more broad application prospects. According to the requirements of new vehicle application, performance and consistency evaluation and state of charge (SOC) prediction of batteries are more and more important, and still a very challenging subject, because it's not easy to measure the separate effect of different internal factors (reaction resistance, contact resistance, and so on) based on the macro parameters obtained from undestroyed testing methods [1].

Although there are not so many reports that cover battery evaluation and prediction work, some articles address key issues, either singly or in some combination. The models usually used for lithium-ion battery evaluation and prediction can be mainly divided into two kinds. First, electrochemical models used for capturing all key behaviors of battery [2−4] can often achieve high accuracy based on the electrochemical theory. They are suitable for understanding the inner electrochemistry reactions in the electrodes and electrolyte [5]. For example, the Peukert equation can associate the battery to an linear system, however, it cannot handle its nonlinear characteristics and it can hardly simulate battery dynamic performance. Normally these equations have a large number of unknown parameters, which leads to huge requirement for memory and computation. They frequently meet over-fitting problems due to their large number of parameters and so on. Hence, electrochemical models are not desirable for actual battery management in electric vehicles. Secondly, equivalent circuit battery models have been investigated especially for vehicle power management and battery management system based on the dynamic characteristics and working principles of the battery by using resistors, capacitors and voltage sources to form a circuit network [6−10]. They are lumped models with few number of parameters. Normally, a large capacitor or an ideal voltage source is used to represent the open-circuit voltage (OCV), and the remainder of the circuit is used to simulate the battery's internal resistance and dynamic effects. Based on the OCV estimate, SOC could be obtained via a lookup table. The equivalent circuit models have been widely used in various types of modeling and simulation battery management systems, although it is not easy to achieve high accuracy.

In this work, a new general battery testing method

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TABLE I Cell type and components[a].

	Components	Version	C^{b}/Ah
LS1	$LiFePO_4$	18650EL	0.9
LS2	$LiFePO_4$	IFR-18650EC	1.3
SS1	Li(NiCoMn)O	INR18650-13Q	1.2
SS2	Li(NiCoMn)O	ICR18650-26F	2.5
SY1	Li(NiCoMn)O	UR18650SA	1.2
SY2	Li(NiCoMn)O	UR18650FM	2.4

[a] LS1: Lishen 1, LS2: Lishen 2, SS1: Samsung 1, SS2: Samsung 2, SY1: Sanyo 1, and SY2: Sanyo 2.
[b] Standard capacity.

is proposed based on the investigations of the thermal property, heat generation behavior and energy loss of batteries, and we discuss the possible rationale behind the experimental design and early data. While we recognize that the datasets we got are still underdeveloped due to recent stage of these measurements, we can see the key fitted parameters (resistance and polarization) of battery are more sensitive, and are closely related to battery performance metrics. The usefulness of these models is compared using validation test data obtained from different cells of the different electrode materials. The advantage of this work, upon completion, will be the testing method used for evaluate performance and consistency and providing more realistic and accurate SOC prediction.

II. EXPERIMENTS

A. Cell components

Several kinds of 18650 Lithium-ion batteries were used to investigate the applicability of the model (Table I). Two types of 18650 cells from three manufacturers were chosen for comparative measurement and analysis, in which number 1 is power type cell and number 2 is energy type cell. All the batteries were chosen from the same batch and produced by Lishen (China), Samsung (Korea), and Sanyo (Japan), respectively.

B. Experiment and theory

All the measurements were carried out under isothermal environment. The charge-discharge tests were carried out with NEWARE BTS-5V/3A type battery tester (China) and MACCOR 4000 battery tester (US) at different current density in different voltage range.

Previous studies showed that the energy balance for batteries consisted of chemical reactions, electrical work, ionic mixing, and heat transfer with surroundings [11−15]. In the condition of neglecting the enthalpy of ionic mixing and assuming the temperature distribution in the cell is uniform, the energy balance function can be written as:

$$\mathrm{d}Q = I^2R\mathrm{d}t + I\eta\mathrm{d}t + IT\left(\frac{\partial E}{\partial T}\right)_p \mathrm{d}t + MC_p\mathrm{d}T \quad (1)$$

where $\mathrm{d}Q$ is the heat-transfer rate between cell and environment, I is the current, R is the resistance (generate Joule heat), η is the polarization voltage (generate polarization heat), $IT\left(\partial E/\partial T\right)_p \mathrm{d}t$ is the reversible electrochemical reaction heat, and C_p is the average heat capacity of the cell.

Based on this multi-parameter function, we built a series of experiments using the following conditions: firstly, kept the testing cells on the constant temperature, so the value of $MC_p(\mathrm{d}T/\mathrm{d}t)$ can be neglected, then because the reversible electrochemical reaction heat had opposite sign during charge and discharge process, so we charged and discharged the batteries using the same current and the same time to counteract the value of $IT\left(\partial E/\partial T\right)_p \mathrm{d}t$. The tests were carried out as the following steps: (i) All the cells were given 1 C charge (default cut-off current is 0.02 C) and discharge current to measure the cells capacities, then the cells were charged to 10% SOC according to their previously obtained capacities. (ii) Rest for 1 h. (iii) Charge the cell at current I_1 for time t_1. Calculate the energy J_{charge} during the charge process. (iv) Rest for 1 h. (v) Discharge the cell at current I_2 for time t_2. Calculate the energy $J_{\mathrm{discharge}}$ during the discharge process.

We ignored the irreversible reaction energy, which was a very small value compared with other energies, considering the cell back to the initial state after the cycle (reversible reaction). The energy balance equation of the cell can be written as below:

$$J_{\mathrm{charge}} + I_1\left(\frac{\partial E}{\partial T}\right)_p Tt_1 = {I_1}^2Rt_1 + \eta I_1t_1 + J_{\mathrm{discharge}} + I_2\left(\frac{\partial E}{\partial T}\right)_p Tt_2 + {I_2}^2Rt_2 + \eta I_2t_2 \quad (2)$$

In the above equation, the left part is the energy absorbed by the cell and the right part is the energy released from the cell. In the testing charge-discharge process, we used the same current and the same time, so Eq.(2) can be rewritten as below:

$$\Delta J = J_{\mathrm{charge}} - J_{\mathrm{discharge}} = 2I^2Rt + 2\eta It \quad (3)$$

where t is the total time of charge-discharge test process (t=t_1=t_2), I is the same current (I=I_1=I_2), and J is the energy loss which can be easily measured by battery testing instrument. So the key parameters remained to be fitted are only R and η.

In order to obtain sufficient data to fit the key parameters, a full factorial experimental plan was developed,

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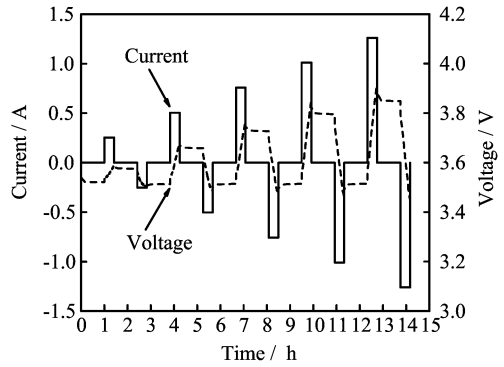

FIG. 1 The charge-discharge current and voltage changing with time (SY1).

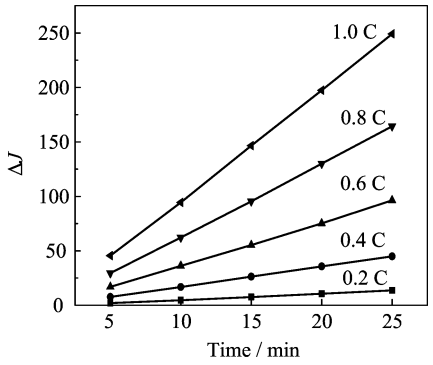

FIG. 2 The energy ΔJ changing with time (SY1).

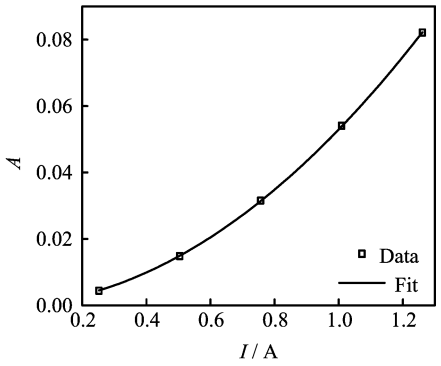

FIG. 3 Fitting curve of A as a function of I (SY1).

which contained five different levels of current (0.2, 0.4, 0.6, 0.8, and 1.0 C) and five different levels of time (5, 10, 15, 20, and 25 min), the current and the voltage change of SY1 during a certain level of time (10 min) are shown in Fig.1. All the measurement process began at 10%SOC and had 1 h rest for the interval.

C. Modeling procedure

It can be seen in Eq.(3) that the different values of charge and discharge energy are determined by two external factors (time and current), and different cells energy values are also determined by two internal factors (R and η). Based on the definition, R is the sum value of similar type of factor inside the cell, which has resistance properties and causes Joule heat generation during charge-discharge process, and η is related with polarization affected by internal factors of the cell, and we assume η is a kind of average parameter which doesn't change with charge-discharge current. Herein, both R and η are the characteristic factors and don't change with current or time. In order to obtain these two important factors, series of measurements were carried out at different current and time according to Eq.(3).

It can be seen in Eq.(3), t has a zeroaxial linear relationship with ΔJ and the slope is $(I^2R+\eta I)$. We measured the changes of energy versus different charge-discharge time (5, 10, 15, 20 and 25 min) in each different current (0.2, 0.4, 0.6, 0.8 and 1.0 C). Figure 2 shows the relationship between ΔJ and t at the same temperature (30 °C). Using straight line can fit five sets of data points very well. We used a linear fitting procedure for five different currents to obtain the parameters of the slope.

Based on the slope data obtained through fitting, we used variable A to stand for the slope, then the model is written as $A=RI^2+\eta I$. I is the only variable. We used a zeroaxial quadratic function curve to fit model $A=RI^2+\eta I$, as shown in Fig.3. Using this method, we can get the exact and relatively constant values of R and η of a particular cell in a dynamic charge-discharge condition, and the values of R and η can reflect some internal properties (reaction resistance, polarization parameter, electrode structure consistency and so on) of the cell, which cannot be easily obtained through the routine tests.

D. Test and verify

In order to verify whether this method and the fitting R and η values were correct, we did the calorimetry measurements which were carried out with HEL PHI-TEC I Adiabatic Reaction Calorimeter (ARC) and PHI-TEC Battery Test Calorimeter (UK). The cell was charged at a certain current (1, 1.5, or 2 C) from 0%SOC to charging cut-off voltage in the adiabatic condition (initial temperature is 30 °C); after the cell back to the initial temperature state, it was discharged at the same current to 0%SOC in the adiabatic condition, and the heat rate of the cell was recorded. The heat rates are shown in Fig.4, which are measured during the cell charge-discharge process.

It can be seen in Fig.4 that the curve trends of heat rate from charge and discharge process of the cell are inverse, because the reversible electrochemical reaction heat is normally opposite during charge and discharge process. We assume that there is an average heat rate

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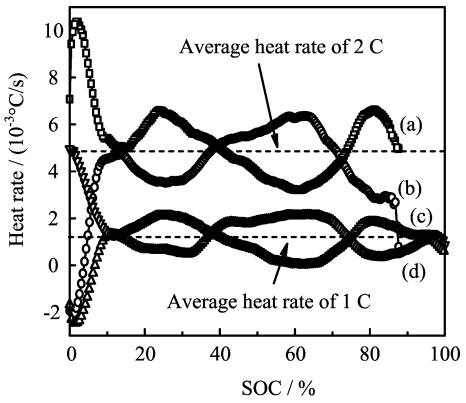

FIG. 4 The curve of charge-discharge heat rate (SY1). (a) Discharge of 2 C, (b) charge of 2 C, (c) Discharge of 1 C, and (d) charge of 1 C.

TABLE II The actual and calculated results of the charge-discharge heat rate of the cell (SY1).

	Average heat rate/(10^{-3}°C/s)		
	1 C	1.5 C	2 C
Actual value	1.2	2.7	4.8
Calculated value	1.3	2.8	4.8

(dotted line in Fig.4), which represents the heat generated from the total resistance and polarization of the cell and doesn't change with SOC. And the cell was tested in adiabatic condition, so all the heat generated from the cell was contributed to the temperature rise, then we can obtain the energy balance function as follows:

$$\frac{I^2R + I\eta}{C_p m} = \frac{\mathrm{d}T}{\mathrm{d}t} \tag{4}$$

where I^2R and the $I\eta$ are the heat generated from the total resistance and polarization of the cell respectively, C_p is the average specific heat capacity of the cell (1.6 J/(g °C)), which can be easily got from the routine thermal meter measurement with ARC, m is the mass of the cell (44 g), and $\mathrm{d}T/\mathrm{d}t$ is the average temperature rise rate. From Eq.(4), we can calculate the value of the average temperature rise rate of the cell (R and η of the cell could be obtained using the previous method) and compared it with actual measured value. The results are listed in Table II, and we used three different charge-discharge current (1, 1.5, and 2 C) to verify the feasibility of this method.

In this process, we neglected the effect of temperature rise on R and η. From the results, we can see that all the actual measured values in different currents have a high consistency with the calculated values which were obtained by the above method. Herein, we can verify that most of the loss energy from the cell during charge-discharge process is caused by the two primary parameters: R and η.

TABLE III Fitted results of different cells.

Version	DC-R/mΩ	R/mΩ			η/mV		
		1	2	3	1	2	3
LS1	18	79	66	67	9.5	9.8	10.4
LS2	38	112	113	104	26.9	28.1	28.0
SS1	20	69	66	70	8.0	7.3	8.5
SS2	46	106	103	106	24.3	25.0	25.8
SY1	21	47	57	54	5.9	6.0	6.3
SY2	38	90	106	90	4.7	3.1	7.7

III. RESULTS AND DISCUSSION

A. The relationship between R-η factors and consistency of cells

Using the above method, different cells (as shown in Table I) were tested to compare the values of R and η, the results are listed in Table III. These tests were all carried out at 30 °C, and three copies for each cell. DC-R was measured through the impedance method with NEWARE BVIR battery impedance tester (1 kHz).

The values of R from the same version of cells indicate that although they all have the same DC-R value, their fitted R values are very different from each other, and the fitted R is much greater than the DC-R. One of the possible reasons is that the fitted R is the sum of all resistance property values inside the cell, so it contains the DC-R and other resistance (reaction resistance and so on), which is not easy to confirm what kind of resistances are inside and measure the effect percentage of different resistances, however using our model, we can get a fitted R factor more closer to the total true value, consequently, we can calculate the Joule heat released from the cell during charge-discharge process based on the fitted R value, which is more accurate than that only based on the DC-R value to obtain Joule heat. It also can be seen that the fitted R values are different, even the three tested cells from the same brand and the same batch, although their DC-R values are highly consistent, since the fitted R also include the internal properties of the cell, which is difficult to keep them exactly the same in the manufacturing process. So we can use this value to evaluate the consistency and the nuances of the cells. The η values don't show big difference between different cells in the same version, which is within the acceptable error range.

From the data of different version cells listed in Table III, it can be seen that most of energy cells have much greater values of R and η than power cells' values (except SY2), which is the same trend as the DC-R change. Smaller values of resistance and polarization can help the power cells to provide higher power capability and decrease the energy loss during high current charge-discharge process, which is consistent with the different requirements of the different kind of cells (power and energy).

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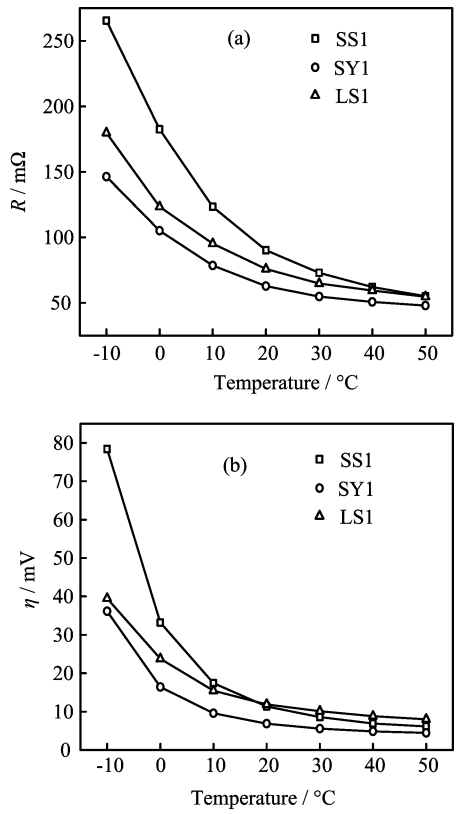

FIG. 5 The changing of (a) R and (b) η with temperature.

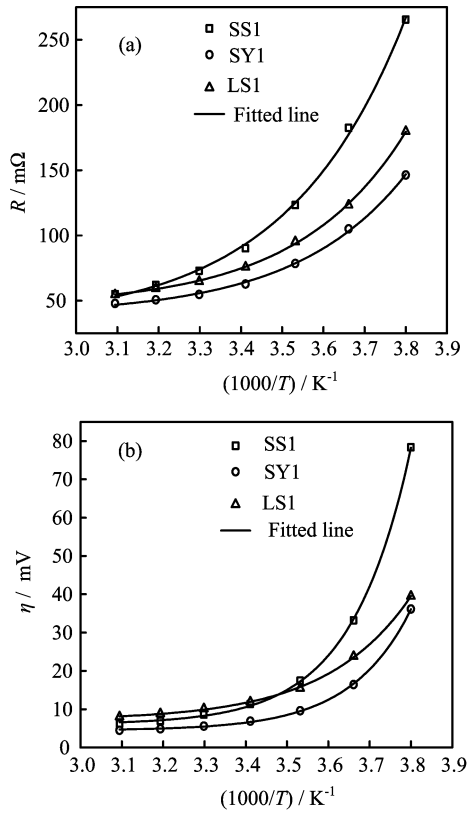

FIG. 6 (a) The changing of (a) R and (b) η with $1000/T$.

B. Effect on temperature to the parameters

A series of measurements have been carried out to investigate the effect of temperature on the parameters. Figure 5 shows how the values of R and η change with temperature.

From Fig.5(a), it can be seen that the value of R increases regularly when the temperature decreases, which is consistent with the normal phenomenon "the resistance of the cells will increase at low temperature". The greater resistances value at lower temperature is one of the reasons for more Joule heat and higher polarization voltage, which is the main impact factor to the performance of the cells at low temperature.

From Fig.5(b), it can be seen that the value of η also increases when the temperature decreases. η stands for the average value of polarization voltage, which will change with SOC during charge-discharge process at different temperature. Although the value of η doesn't change as regularly as R, we can still obtain the changing trend of η, which also indicate that lower η value is better to obtain higher cell performance.

In order to obtain the function of R and η relating to temperature respectively, we developed a general model, which is written as follows:

$$\begin{aligned} R &= R_0 + B_1 \exp\left(\frac{1000}{b_1 T}\right) \\ \eta &= \eta_0 + B_2 \exp\left(\frac{1000}{b_2 T}\right) \end{aligned} \tag{5}$$

where R_0 and η_0 are the resistance and the polarization voltage of the cell when T is infinite, which are the intrinsic factors of the cell and don't change with temperature. The value of B is the increasing rate of R and η with the varying of temperature, which is also a kind of cell internal factors. We fixed the values of b to 0.27 and 0.16, respectively for R and η, in order to obtain a more significant R_0 (η_0) and B. From Fig.6, it can be seen that using Eq.(5) could fit the data of R and η very well. The values of the fitted parameters of Eq.(5) are shown in Table IV.

The complete model function is written as below, which can account for various energy losses of the cell during charge-discharge process at different temperatures and is a general model for most of lithium-ion batteries:

$$\begin{aligned} \Delta J = I^2 \left[R_0 + B_1 \exp\left(\frac{1000}{0.27T}\right)\right] t + \\ \left[\eta_0 + B_2 \exp\left(\frac{1000}{0.16T}\right)\right] It \end{aligned} \tag{6}$$

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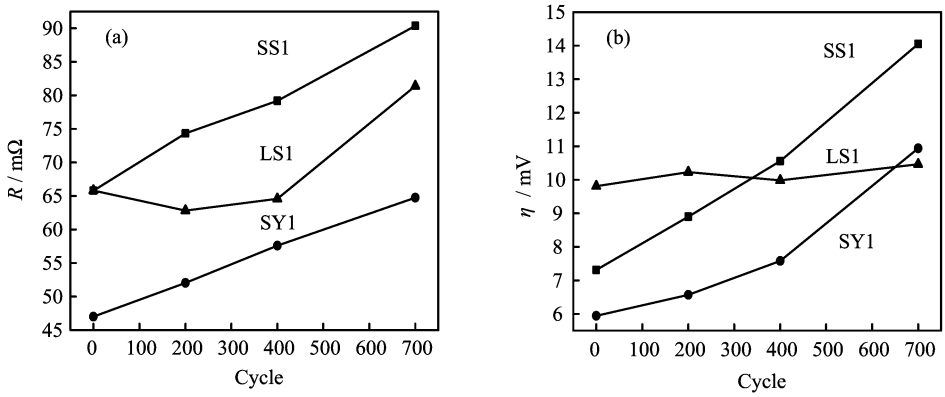

FIG. 7 The changing of (a) R and (b) η with cycle number.

TABLE IV Fitted results of R_0, η_0, B, and b of different cells (b_1=0.27, b_2=0.16).

Version	R_0/mΩ	$B_1\times10^{-5}$	η_0/mV	$B_2\times10^{-9}$
LS1	44	10	8.8	1.5
SS1	38	18	4.7	3.5
SY1	39	8.3	4.0	1.5

This general model includes four factors R_0, B_1, η_0, B_2, which can be applied in the temperature region of −10 °C to 50 °C at least. In the model function, R_0 is an intrinsic factor of the cell that reflects the internal resistance properties of the cell and B_1 is the temperature coefficient of R. η_0 is also an intrinsic factor of the cell that reflects the internal polarization properties of the cell and B_2 is the temperature coefficient of η. The research on the applications of R_0, B_1, η_0, B_2 in the widely evaluation of consistency of different cells and the calculation of the energy loss during charge-discharge process is still ongoing.

C. Effect of cycles on the parameter

To identify R and η parameters change over the whole performance life, series of tests has been carried out in order to investigate the effect of the cycle numbers on the parameters, as shown in Fig.7.

All the performance tests and cycle tests were carried out at 30 °C. We used a 1 C charge-discharge current for the cycle test. As seen in Fig.7, parameters were measured and fitted at the cycle numbers of 0 (initial point), 200, 400, and 700. Both R and η had a trend of increase for the SS1 and the SY1, the clear exception was the data of LS1 (which don't have a regular trend for both parameters). Because the R and η were the key parameters for the Eq.(3) which reflected the energy loss during charge-discharge process, so we related the 1 C discharge energy at different cycles (Fig.8) to the change of these parameters. Clearly, the SS1 and

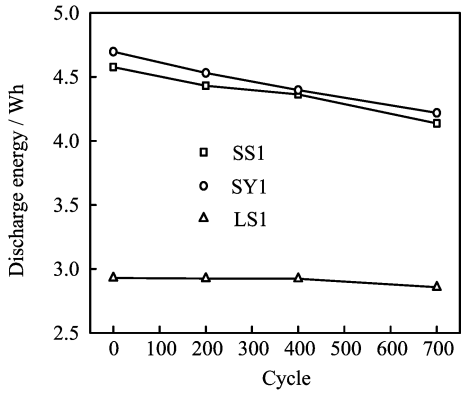

FIG. 8 The changing of discharge energy with cycle number.

SY1 battery had an obviously decrease trend for their discharge energy after cycles, which could be explained by the increase of R and η. R and η for LS1 didn't have an obviously change during the first 400 cycles and after 700 cycles the R value began to increase, which could be explained by Fig.8 in that the discharge energy of LS1 changed little during first 400 cycles and then decreased after 700 cycles. The further investigation (manufacture procedure, electrode materials, formula and so on) on the different performance of LS1 is still ongoing.

Normally, the trend of R is increasing with cycling measurement (except the data of LS1), in view of R factor which is very sensitive and comprehensive to reflect the internal situation and changes of cells, if the trend of R is becoming decreasing on one particular cycle, which might indicate that some important changes (micro short circuit and so on) appear inside the cells, and it's an important signal to the cell security.

D. Battery surplus energy prediction

Based on the above model, R and η parameters can be used to calculate the energy loss of the batteries dur-

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TABLE V Surplus energy prediction at constant current in different temperatures*.

Version	30 °C			0 °C		
	E_M*	E_C*	Error/%	E_M*	E_C*	Error/%
LS2	4.7378	4.7339	−0.08	2.9843	3.0324	1.61
SY1	4.4086	4.4133	0.11	3.5165	3.5796	1.79

* E_M and E_C are measured and calculate value, which are in units of Wh.

TABLE VI Surplus energy prediction at pulse current in different temperatures.

Version	30 °C			0 °C		
	E_M*	E_C*	Error/%	E_M*	E_C*	Error/%
LS2	1.2461	1.2400	−0.49	1.2089	1.2100	0.09
SY1	1.0842	1.0791	−0.47	1.0170	1.0426	2.52

* E_M and E_C are measured and calculate value, which are in units of Wh.

ing charge-discharge process. With these data we can predict the surplus energy after the battery discharged at a certain current and a certain time. The method is as follows:

$$E_{\text{discharge}} = E_{\text{charge}} - (I^2_{\text{charge}} R t_{\text{charge}} + I_{\text{charge}} \eta t_{\text{charge}}) - (I^2_{\text{discharge}} R t_{\text{discharge}} + I_{\text{discharge}} \eta t_{\text{discharge}}) \quad (7)$$

where $E_{\text{discharge}}$ is the remaining energy of the cell, E_{charge} is the charging energy into the cell which could be got from the battery testing instrument, $(I^2_{\text{charge}} R t_{\text{charge}} + I_{\text{charge}} \eta t_{\text{charge}})$ was the energy loss during charge process, and $(I^2_{\text{discharge}} R t_{\text{discharge}} + I_{\text{discharge}} \eta t_{\text{discharge}})$ is the energy loss during discharge process. So the energy input to the cell subtracts the energy loss to be the energy which could output from the cell. We used this thermal property based method to predict surplus energy of the cells, which were charged and discharged at 1 C constant current at two different temperatures (0 and 30 °C). The results are listed in Table V.

From the results, we can see that the surplus energy calculated at 30 °C was very closed to the measured value, and at 0 °C the error was less than 2%. Then we did the measurements, when the cells were discharged at different pulse current. Firstly, the cells were charged to 80%SOC (charge energy was measured), then the cells were discharged at 2 C current (36 S) and 0.5 C current (288 S) to about 30%SOC, at last the surplus energy was measured at 1 C discharge current to contrast with the calculated value. The values were also tested at two different temperatures (0 and 30 °C). The results are listed in Table VI.

Under different pulse current discharge condition, this method also has a high accuracy. So we can use this thermal property and energy loss based prediction method to calculate the surplus energy which could be output from the cells. Using the results got from this method, we can get more information (effective percentage of output energy and so on) to evaluate the performance of the cells, which is important to battery management system, and the results also verified the rationality of our battery evaluation research based on thermal property and heat generation behavior of battery.

IV. CONCLUSION

We studied thermal property and heat generation behavior in lithium-ion batteries charge-discharge process. According to energy analysis, a general multi-parameter model (Eq.(3)) was developed and its applicability was investigated on a series of lithium-ion batteries with different capacities. The results show that the theoretical model function can be in good agreement with the actual situation. The applicability of the model function was also increased to a wide range of temperature from −10 °C to 50 °C, which is more appropriate for electric vehicles battery environment. The input and output energies of the cell during charge-discharge process are the key measurement parameters. According to the relationship between energy, time and current, the cell resistance R and polarization voltage η can be obtained through the curve fitting method. The value of R is a total resistance of the cell including DC-resistance and other resistance-feature factors, which can account for the generation of Joule heat. η reflects the internal polarization properties of the cell. Consequently, R and η are both very important factors to the evaluation of the performance and the consistency of the cells. Further, a general semi-empirical model Eq.(6) was developed to describe the changes of R and η with the varying of temperature, which can be used to define the exothermal behavior of the cells at different temperatures. Then, the relationship of R and η with cycle numbers was discussed to verify that these two parameters were the key factors to the energy loss in batteries' charge-discharge process. We built a new SOC prediction method based on this model to predict surplus energy of the batteries. The results were closed to actual measured values at different temperatures and different type of testing currents (constant and pulse). Some related further research is still ongoing.

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